

effect on ϕ , and do not change our other conclusions.

In summary, the data for χ_c , the overall shapes of the phase boundaries, and the experimentally derived ϕ support the conclusion that the para-ferro-fan triple point in MnP is a LP.

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^(a)Present address: Instituto de Física, Universidade de São Paulo, C.P. 20516, São Paulo, Brazil.

^(b)Present address: Center for Space Research, Massachusetts Institute of Technology, Cambridge, Mass. 02139.

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Observation of Solid ³He Ordering at Melting Pressures in High Magnetic Fields

H. Godfrin, G. Frossati, A. S. Greenberg,^(a) B. Hébral, and D. Thoulouze

Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique,
F-38042 Grenoble, France

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With use of compressional cooling, ordering in solid ³He is observed and identified by a null in the pressurization rate for reversible compressions in magnetic fields up to 7.2 T. At this field the ordering is above 3 mK. Entropies at T_A deduced from $(P_{A_2} - P_{A_1}) / (T_{A_2} - T_{A_1})$ are shown to be lower than expected from the various models. Irreversible magnetic heating at high fields which limits the effective cooling power is also observed.

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A rapid decrease in the entropy of solid ³He at 1.1 mK was first identified as the nuclear ordering transition by Halperin *et al.*¹ The effect of a moderate magnetic field on the ordering has been studied by several groups,^{2,3} stimulating a variety of theoretical models: multiple spin exchange,⁴ defect-induced⁵ or polaron-induced ordering,⁶ and

spin-glass.⁷ In this Letter, we report pressure measurements in magnetic fields up to 7.2 T as well as results of entropy measurements at the temperature 2.75 mK.

The compressional cell,⁸ including the compressional membrane, was constructed entirely from plastic. The liquid volume was 2.1 cm³.

Capacitance gauges measured the ^3He pressure and volume. The entire assembly was placed inside the mixing chamber of a dilution refrigerator that maintained steady-state temperatures between 2.4 and 3 mK depending on magnetic field. This proved to be important in our studies since most of the solid was formed at, or very close to, the ordering temperature.

We will use in this paper an experimental definition of "ordering," which does not imply the existence of a long-range-order transition, but implies at least a substantial decrease of the entropy. We define ordering as the point where dP/dt is almost zero. This means that the cooling power balances the heat leaks (about 10^{-10} W), as well as possible irreversibilities which will be discussed later.

We show that the entropy drops rapidly at ordering, the melting curve becoming essentially flat. Therefore, this experimental definition should give reliable values for the ordering pressures; the corresponding ordering temperatures used to establish the H - T magnetic-phase diagram are inferred in a way which will be explained in the text.

In Fig. 1, we show the ordering pressure, as well as the superfluid ^3He transitions A_1 and A_2 , as a function of magnetic field. The ordering pressure decreases linearly with H , for $H > 0.5$ T, with $dP/dH = -3.4$ MPa/T at the ordering temperatures. Since $dP/dH \sim \Delta M/\Delta V$, at least at high fields where the ordering temperature does not depend strongly on H , we conclude that the magnetization in the high-field ordered phase is close to 70% of the saturation magnetization. This value essentially corresponds to the normal antiferromagnetic behavior predicted in this H - T range by the multiple-spin-exchange model.^{4,9}

The pressure splitting of the A transition previously measured at lower fields can be described as essentially linear, although deviations from linearity at 2.8 T, $P_{A_2} - P_{A_1}$ increasing somewhat faster, were reported by Schuberth, Bakalyar, and Adams.³ On the contrary, here we observe $\Delta P = P_{A_2} - P_{A_1}$ to reach a maximum, then remain roughly constant for $2 < H < 4$ T. The temperature splitting was reported to be $64 \mu\text{K}/\text{T}$ at low fields,¹⁰ and $74 \mu\text{K}/\text{T}$ at 2.8 T.³ Present theories predict a linear temperature behavior up to high fields.¹⁰ The roughly constant pressure splitting thus reflects the decrease of the melting curve slope near the ordering for $H > 2$ T, as discussed later on.

Direct measurements of the detailed splitting

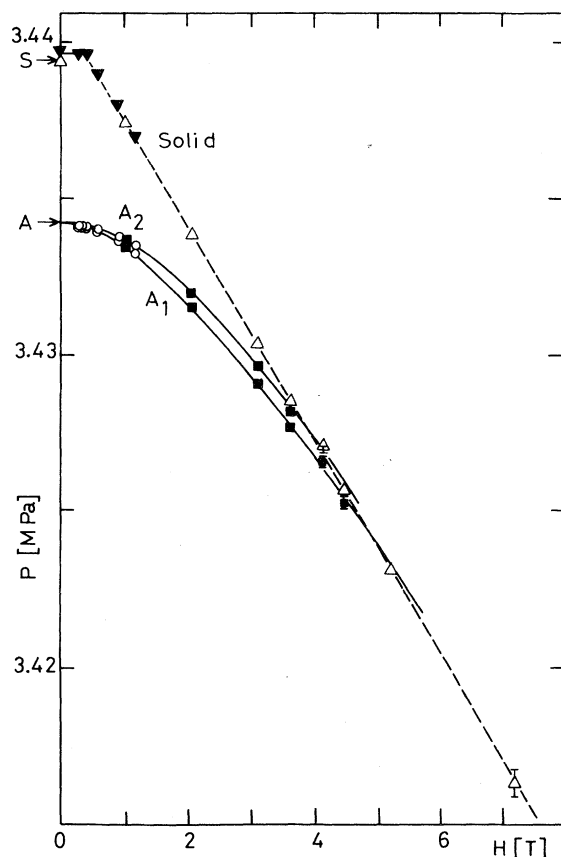


FIG. 1. Characteristic pressures in a Pomeranchuk cell vs magnetic field. Equilibrium maximum pressure: S, Ref. 1; inverted solid triangles, Ref. 2; and open triangles, this work. A_1 and A_2 superfluid transitions of liquid ^3He : A, Ref. 1; open circles, Ref. 2; and solid squares, this work.

are called for, but it will be sufficient for the purposes of this experiment to *assume* a linear splitting of $\sim 64 \mu\text{K}/\text{T}$,^{10,3} and that a 2.75-mK isotherm can be imagined to run between the A_1 and A_2 curves. It can be seen in Fig. 1 that at 4.2 T the ordering is at the same pressure as A_2 , $T \sim 2.6$ mK, while at 5.0 T it appears at the A_1 transition, $T \sim 2.9$ mK. For higher fields, ordered solid ^3He is no longer formed from superfluid, but from normal liquid ^3He .

Of primary importance to both theories and experiments is the magnetic (H , T) phase diagram. In our experiments, the ordering temperatures can be estimated from the ordering pressures of Fig. 1 by use of the melting curves^{2,3} for fields below 3 T, and the linearity of the A temperature splitting as discussed before for $3 \text{ T} < H < 5 \text{ T}$. The ordering temperatures increase with the magnetic field, as predicted by the multiple-spin-ex-

change model⁴ and the vacancy-ferromagnet model.⁶ In Fig. 2 our results have been compared to the results of the multiple-spin-exchange model,⁴ which to date offers the most detailed phase diagram in high magnetic fields. It is seen that the observed ordering and the theoretical boundary (a line of second-order transitions) have the same general field dependence except for what appears to be a constant temperature shift. However, the difference is larger than appears in Fig. 2 since our experimental definition of ordering results in consistently lower experimental values of ordering temperatures. In fact, the magnitude of the entropy near the experimentally defined ordering is $<0.2R$, according to entropy measurements at low fields^{2,3} and cooling-power experiments at 7.2 T.

The pressures of the A_1 and A_2 transitions of the liquid provide an independent confirmation of this large entropy reduction in the solid. In Fig. 3, we show values of the solid entropy at constant temperature, T_A , deduced from a linear A splitting with use of the Clausius-Clapeyron equation:

$$\begin{aligned} dP/dT &\sim (P_{A_2} - P_{A_1}) / (T_{A_2} - T_{A_1}) \\ &\sim (S_L - S_S) / (V_L - V_S). \end{aligned}$$

For $0 < H < 2$ T, the entropy is almost field independent, and close to the zero-field value of Ref.

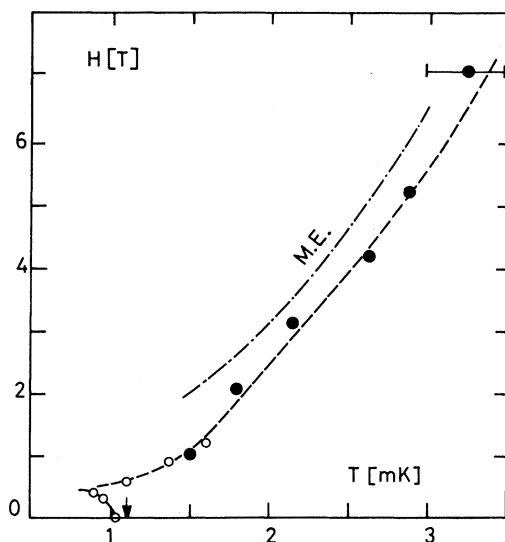


FIG. 2. Magnetic field vs temperature phase diagram of solid ^3He . Arrow, Ref. 1; open circles, Ref. 2; and solid circles, experimentally defined ordering temperatures, this work (see text). Curve ME is from multiple-exchange model (Ref. 4).

1. For $H > 2$ T, $S(H, T_A)$ decreases, with values close to that of free spins. The large antiferromagnetic interactions responsible for the zero-field ordering ($J \sim -0.75$ mK) can be taken into account in a mean-field calculation; the results are shown in Fig. 3 and can be approximated by $S = \ln 2 - 0.5[\mu H/k(T + \Theta)]^2$ with $\Theta = 3$ mK. For all fields the measured entropy at T_A is much lower than the calculated values.

By bringing the cell into equilibrium with the mixing chamber, which contained a field-independent capacitance thermometer,⁸ we obtained the melting curve at 7.2 T, and consequently the solid ^3He entropy by the Clausius-Clapeyron equation. The results, summarized in Fig. 4, show that $S(T, H = 7.2$ T) is close to the entropy of a paramagnet ($H \sim 7$ T). Again, no agreement can be found with a mean-field calculation with $J = -0.75$ mK, as was also observed by Johnson, Rapp, and Wheatley¹¹ at 6.36 T.

The large departure from the molecular-field approximation with antiferromagnetic interactions indicates a "ferromagnetic" tendency, also observed in low-field thermodynamic measurements. It may also show the onset of an ordering

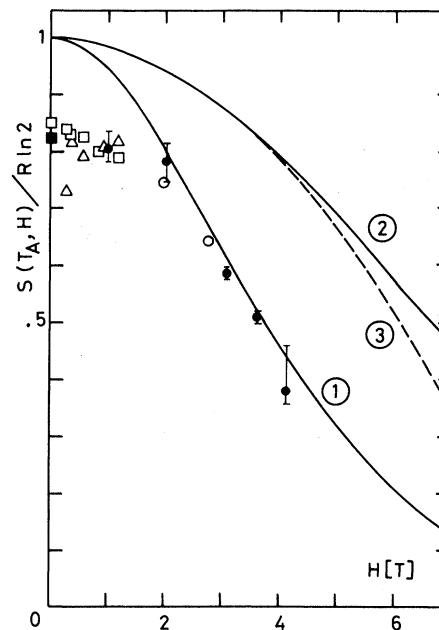


FIG. 3. Entropies of solid ^3He at T_A vs magnetic field. Solid squares, Ref. 1; open squares, Ref. 2; open circles, Ref. 3; open triangles, calculated from the data of Ref. 2 following the procedure described in the text; solid circles, this work. Curve 1, free spins; curve 2, molecular field, $\Theta = 3$ mK; curve 3, $S = \ln 2 - 0.5[\mu H/k(T + \Theta)]^2$, $\Theta = 3$ mK.

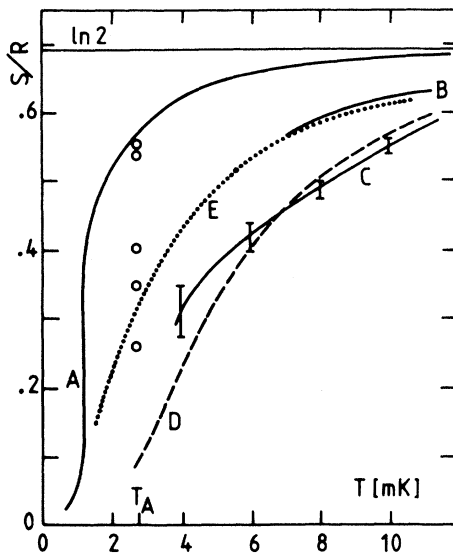


FIG. 4. Entropy of solid ^3He vs temperature for different magnetic fields. Curve A, Ref. 1 ($H=0$); curve B, Ref. 10 ($H=6.36$ T); curve C, this work ($H=7.2$ T); curve D, free spins ($H=7$ T); curve E, mean-field calculation, $\Theta=3$ mK ($H=7$ T), see text. Open circles are our data ($H=1.04, 2.09, 3.13, 3.65,$ and 4.18 T).

with a well-defined transition, as observed^{1,2} for fields up to 1.2 T. For $H < 4$ T, a comparison of the magnitude of the entropies measured at T_A and at the "ordering" imply a substantial decrease of the entropy in a temperature interval of less than 1 mK. This suggests an ordering transition. More extensive measurements are in progress to determine the nature of the ordering.

One has to be very cautious about the meaning of thermodynamical measurements in high fields, because of the possible existence of long time constants between the different energy reservoirs of solid ^3He .¹² Therefore, in a Pomeranchuk cell, consideration has to be given to the rate of conversion of liquid to solid and the dynamical effects associated with the solid formation.¹³ That such effects are important has been pointed out by Delrieu⁹ and Yu and Anderson.¹⁴ They attribute the "backstep" in the pressurization curve observed by Schuberth, Bakalyar, and Adams³ to a change in the spin relaxation mechanisms going from normal to superfluid ^3He . We can similarly observe the backstep for irreversible compressions up to fields of 4 T, but at fields higher than 5 T, where the solid is formed from the normal Fermi liquid, no backstep was observed. This supports the interpretation of the backstep as being an effect due to the liquid.

A related effect was produced by rapidly increasing the compression rate from the point $dP/dt=0$. We observe that the pressure once again increased, and then rapidly decreased even though the compression was continued at the same high rate ($>3\%$ of solid/min). At 7 T, pressures as high as the B' transition pressure in zero field could be obtained. With such a rapid compression the liquid cannot supply the solid with the proper polarization even in the superfluid phase. The solid is then formed with a reduced magnetization, and the pressure is that of a melting curve at lower effective external fields. This is similar to the initial part of the backstep. When the solid magnetization relaxes by ΔM , an amount of energy $\Delta Q \sim H_0 \Delta M$ is released and the pressure decreases, because of both the irreversible heating and the return to the equilibrium melting curve which is at a lower pressure. The Pomeranchuk cooling will in fact be halted when $T\Delta S = H\Delta M$, as pointed out by Delrieu.⁹

This behavior is characteristic of a highly irreversible compression. For very slow compressions ($<3\%$ of solid/h), we have verified that the maximum pressure is independent of the compression rate; the pressure of the A_1 feature is the same for compression or decompression; and the backstep can hardly be recognized. If the compression is stopped, the pressure decreases only slightly from the maximum pressure. We have considered these experiments as "reversible compressions" and the measured pressures as the equilibrium values.

We have considered the possibility that the observed ordering could be just an effect of magnitude relaxation heating. However, this is inconsistent with the observed field dependence of the experimental results shown in Fig. 2; at fields below 2 T even a 100% magnetic irreversibility is not sufficient to stop the cooling unless the entropy is seriously reduced; at higher fields a large irreversibility ($\sim 50\%$) would be needed. This is in contradiction with the results on maximum pressure versus compression rate and direct NMR measurements of the magnetization at 7.2 T which are in progress.

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^(a)Address during 1980: Service de Physique des Solides et de Résonance Magnétique—Commissariat à l'Energie Atomique, Centre d'Etudes Nucléaires de Saclay, Orme des Merisiers, B.P. 2, F-91190 Gif-sur-Yvette, France.

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Solid ³He Magnetic Phase Diagram from Static Magnetization Measurements

T. C. Prewitt and J. M. Goodkind

University of California, San Diego, La Jolla, California 92093

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The magnetization of solid ³He has been measured at a density of 24.2 cm³/mole for fields between 0.8 and 5.8 kG, and for temperatures from 0.55 to 2.0 mK. The measurements reveal the magnetic properties of the phases previously observed by Kummer, Mueller, and Adams, and are in qualitative agreement with the phase lines obtained by them. A new second-order transition has been observed for a narrow range of fields above 4.1 kG.

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In recent years several experiments have shown that solid ³He undergoes a first-order magnetic phase transition at 1.0 mK.¹⁻⁶ Kummer, Mueller, and Adams² have found that in a magnetic field this transition remains first order up to 4.1 kG and then changes character to a second-order transition for higher fields. In order to study this unanticipated result, we have measured the static magnetization, *M*, versus temperature in fields, *H*, from 0.8 to 5.8 kG, and at temperatures down to 0.55 mK. Our measurements reveal the behavior of the magnetization of the previously observed phases, and also show a new transition which exists in a narrow range of fields above 4.2 kG.

A sample of solid ³He was cooled by means of adiabatic demagnetization of a bundle of copper

wires. The ³He was contained within a sintered silver sponge with a surface area of 1.0 m² and an available volume of 0.1 cm³. A small portion of the ³He (roughly 10%) occupied a region outside the silver sponge which was left as a consequence of the sponge fabrication. This region had a typical thickness of 0.1 mm. Magnetic fields were applied to the ³He by trapping the required current in a superconducting solenoid wound directly onto the sample container. Accuracy in the setting of magnetic fields was estimated to be 3%. The field was stabilized against drifts by a thin niobium-titanium cylinder between the ³He and the solenoid. The ³He magnetization was sensed by a SQUID magnetometer connected to the sample container by a superconducting transformer. Because of the poor thermal conductiv-