FREEZING OF MAGNETIZED He^3 AT LOW TEMPERATURES

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Cooling by adiabatic freezing of He^3 eventually comes to a dead end because the solid transforms into an antiferromagnet. It is shown that in the presence of a magnetic field the final temperature can be made arbitrarily low and is limited only by the starting temperature of the freezing process.

We recently estimated[1] that the lowest temperature $T_{f,\mu}$ attainable by the adiabatic freezing[2] of He^3 was about 0.5 mK. Since this process now appears to be a useful method for producing very low temperatures, it seemed of interest to study mechanisms whereby $T_{f,\mu}$ might be extended to significantly lower temperatures. We present briefly the results of our study which will be treated in detail in another journal.

The limiting temperature $T_{f,\mu}$ is closely associated with the antiferromagnetic transition temperature T_c of solid He^3. At the upper approaches of T_c, rapid spin ordering occurs in the solid, causing the entropy available for cooling to decrease drastically. Our problem is then one of finding a practical way of lowering T_c.

The spin ordering may be accounted for by exchange coupling between a given nuclear spin and its nearest-neighbor spins of opposite direction. If the exchange potential energy per atom is denoted by $\frac{1}{2}|J|$, then in general the spin-ordering temperature is given by

$$T_c = \nu z|J|/2k, \tag{1}$$

where k is the Boltzmann constant, z is the crystal coordination number, and ν is a positive number of the order unity. In a spin-$\frac{1}{2}$ antiferromagnet, such as bcc He^3, the couplings tend to lock the sublattices of the two spins in opposite directions. In the region $T<T_c$, the total spin of the system and the magnetic moment disappear because of mutual compensation of the sublattices. The situation, however, is changed radically by application of a uniform magnetic field parallel to the spin direction of one of the sublattices.[3] This produces preferential magnetizations σ_+ parallel to the direction of the field and σ_- opposite to it such that $|\sigma_+| \geq |\sigma_-|$. The external field transforms the crystal coordination number z into a spin coordination number $z(H)$ which depends on the field strength. Since the coordinated number $z(0)$ of opposite or (−) spins of a given (+) sublattice spin is decreased by the alignment of the field in the (+) direction, then

$$z(H) \leq z(0). \tag{2}$$

When H reaches a value

$$H_c \sim kT_c(0)/\mu, \tag{3}$$

where μ is the magnetic moment per spin, the $z(H)$ spins on the (−) sublattice are exhausted and the system ceases to be an antiferromagnet. At field strengths $H \leq H_c$, the antiferromagnetic transition temperature is lowered. With Eqs. (1) and (2) we have

$$\begin{aligned} T_c(H) &\simeq \nu z(H)|J|/2k \\ &= [z(H)/z(0)]T_c(0) \\ &\leq T_c(0), \end{aligned} \tag{4}$$

where $T_c(H) \to 0$ as $H \to H_c$ and $z(H_c) \to 0$. The scalar dependence of T_c on H is parabolic, at least as $H \to 0$. In solid He^3 at melting,[1] $T_c(0) \approx 2$ mK which, with the experimental value of μ, yields $H_c \approx 27$ kG.

Because of the cubic magnetic symmetry of He^3, the above calculation changes only slightly for various models that may be chosen to represent the solid. In general this is not true of electronic or ionic antiferromagnets which exhibit structural magnetic asymmetries and other anisotropies. These cause the antiferromagnetic phase to split into two subdivisions which would produce along $T_c(H)$ a triple point where the paramagnetic and two spin-ordered subregions are in equilibrium.

In the absence of a magnetic field, we may represent the He^3 melting anomaly by the temperature derivative of the melting pressure $P_m(T)$. From thermodynamics,

$$\frac{dP_m(T)}{dT} = \frac{[S_l(T) - S_s(T)]}{[V_l(T) - V_s(T)]}, \tag{5}$$

where the subscripts l and s of the molar entropies and volumes denote equilibrium liquid and solid, respectively. The melting anomaly starts at T_μ, where S_l equals S_s and $P_m(T)$ has a minimum; it ends at $T_{f,\mu}$, where $P_m(T)$ has a maxi-

mum. Since $T_{f,\mu} < T_c(0)$, the anomaly terminates when the freezing solid is in its spin-ordered state. At $T < T_c(0)$ or $T < T_c(H)$,

$$S_s(T,H) \geq S_s(T, H \to 0),$$

and

$$(\partial S_s/\partial H)_T \geq 0. \tag{6}$$

Isothermal magnetization of an antiferromagnet is accompanied by an entropy increase because of the asymmetry introduced by the two unequal sublattice magnetizations σ_+ and σ_-. For material in a field, the melting-line branches $P_m(T, H = \text{const})$ generate the melting surface $P_m(T,H)$. For every value of $H = \text{const}$, there is a melting line such that

$$\frac{dP_m(T,H)}{dT} = \frac{[S_l(T) - S_s(T,H)]}{[V_l(T) - V_s(T)]}, \tag{7}$$

neglecting[4] the extremely small changes in entropy of the liquid on magnetization, and in molar volume of the solid spin system on magnetization. At $T < T_c(0)$ or $T < T_c(H)$, Eq. (6) shows that the melting anomaly is enhanced in the presence of a field since $|dP_m(T,H)/dT| > |dP_m(T)/dT|$. This is another way of demonstrating the lowering of $T_c(0)$. The effect of the field on the melting-curve minimum at $T_\mu \sim 0.3$ K is, however, quite negligible. With $S_s(T,H) > S_s(T, H = 0)$, and with both functions being of monotonic variation in T, $T < T_c(0)$, and $S_l(T)$ unchanged, the right-hand side of Eq. (7) can only vanish at $T_{f,\mu}(H) < T_{f,\mu}(0)$. Thus the final temperature $T_{f,\mu}(H)$ accessible by adiabatic freezing of He^3 in the presence of a magnetic field $H < H_c$ is lower than the zero-field value $T_{f,\mu}(0)$.

To illustrate quantitatively the adiabatic freezing in the presence of the magnetic field, it is useful to consider one particular line $P_{M,c}(T,H)$ of the melting surface. This critical melting line refers to those final states of the all-solid sample which are along the critical transition line $T_c(H)$. One finds here $dP_{M,c}/dT \leq 0$, the equality occurring only at absolute zero. By Eq. (7), this melting-pressure branch increases monotonically with decreasing temperatures toward its limit value at absolute zero. With $S_{s,c}(T,H) \geq S_s(T, H = 0)$, which is another expression of the negative temperature derivative of $P_{M,c}(T,H)$, final states of the all-solid sample along $T_c(H)$ enable one, in priniciple, to reach arbitrarily low temperatures.

Shown in Fig. 1 are two sets of final temperatures, indicating the effect of external fields.

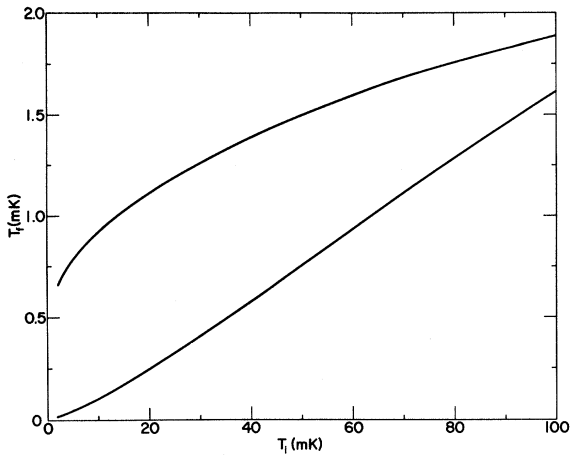

FIG. 1. The effect of an applied uniform magnetic field on the production of very low temperatures T_f by adiabatic freezing of He^3 as a function of the initial temperature T_i of the starting all-liquid sample. The lower curve gives those $T_f(T_i, H)$ values which are associated with all-solid final states along the critical spin-ordering transition line $T_c(H)$. The upper curve refers to cooling in the absence of a magnetic field.

The upper curve is for $T_f(T_i, H = 0)$ obtained earlier,[1] and is to be compared with $T_f(T_i, H)$ shown as the lower curve. The lowest final temperatures are limited by the starting temperatures T_i of the all-liquid sample. At the present time, experimental demonstration of the effect may require measurement of the temperatures reached in adiabatic freezing in the absence and presence of the field, starting with identical initial states of the all-liquid sample and forming the all-solid samples of the same entropy. The measurements of the melting-pressure branches $P_M(T,H)$, at constant field strengths, or differences $P_M(T_1,H) - P_M(T_2,H)$ along these branches, would complete the verification of the predicted melting properties of He^3 in the presence of a magnetic field.

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[1] L. Goldstein, Phys. Rev. 188, 349 (1969), and 159, 120 (1967).

[2] This process, with its estimated high starting temperature, was predicted by I. Pomeranchuk, Zh. Eksp. Teor. Fiz. 20, 1919 (1950); its final low-temperature end due to the probably antiferromagnetic spin ordering of solid He^3 at melting was predicted by H. Primakoff, Bull. Amer. Phys. Soc. 2, 63 (1957).

[3] The asymptotic two-sublattice-model approach to spin-$\frac{1}{2}$ antiferromagnetic systems, on the basis of the molecular-field-theory formalism, is due to J. H. Van Vleck, J. Chem. Phys. 9, 85 (1941), and to C. G. B. Garrett, *ibid.* 19, 1154 (1951).

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