Phenomenological Approach to Paramagnetic Liquid He^3 at Very Low Temperatures*

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Magnetic-susceptibility measurements around the apparent very low transition temperatures of liquid He^3 will be shown to be indicative of the role played by the spin system in the transformation process. This phenomenon is found to be depressed toward lower temperatures on application of external, constant, and uniform magnetic fields, with the ordered modification of the liquid confined to a limited region of its magnetic phase diagram.

In an initial treatment of various equilibrium thermal properties of liquid He^3, we have been led[1] to define the mean number of its thermally excited spins $N_\sigma(T)$ at temperature T along isochores of volume V or isobars of pressure p, of a system formed by N atoms. Omitting the state coordinates, V or p, the fractional number of excited spins was shown to be given[1] by

$$N_\sigma(T)/N=\chi(T)/\chi_0(T), \tag{1}$$

where χ is the actual paramagnetic susceptibility of the system and χ_0 is the susceptibility it would have in the indicated equilibrium state if it were an ideal Langevin-Brillouin[2] spin-$\frac{1}{2}$ paramagnet. The latter reduces here to

$$\chi_0(T, V)=N\mu^2/VkT, \tag{2}$$

μ standing for the magnitude of the elementary magnetic dipole moment. The relation (1) thus pictures the nonideal character of the paramagnetic system as if the magnetization, at low-field strengths, involved only a reduced number $N_\sigma(T)$ of the total number N of the spins of the whole system. As a result of the interactions within the system, the remaining $(N-N_\sigma)$ atoms had their spins blocked, escaping thus the low field-strength magnetization. Fermi statistics expected[3] to affect the behavior of liquid He^3 yields, at low-enough temperatures,[1,4]

$$\lim_{T\,\text{small}}\frac{N_\sigma(T)}{N}=\frac{3}{2}\left(\frac{T}{T_0}\right)\left[1-\sum_{\lambda=1}p_{2\lambda}\left(\frac{T}{T_0}\right)^{2\lambda}\right], \tag{3}$$

where $T_0(V)$, for instance, is a characteristic temperature of the spin system along an isochore V, and the $p_{2\lambda}$'s are numerical coefficients. The formal definition of T_0 is phenomenological[1] through the observable limiting constant Pauli-type susceptibility χ_P at very low temperatures, or

$$T_0(V)=\frac{3}{2}N\mu^2/Vk\chi_P. \tag{4}$$

The mean spin-excitation numbers $N_\sigma(T)$ led one to associate with them a component entropy of spin disorder[1] through

$$\frac{S_\sigma(T)}{Nk}=(\ln 2)\left(\frac{N_\sigma(T)}{N}\right)=(\ln 2)\left(\frac{\chi(T)}{\chi_0(T)}\right), \tag{5}$$

which in turn defines the component spin heat capacity $C_\sigma(T)$ as well as other component derivative thermal properties of the system[5] throughout the diagram of state of liquid He^3.

The above approach explained[1] the heat-capacity plateau observed in liquid He^3 at $T\gtrsim 0.30-0.40$ K. It has led to recognize the universal character[6] of the susceptibility ratio, or the mean fractional number of thermally excited spins by Eq. (1), in that either one of these properties was a unique function of the phenomenological reduced temperature $[T/T_0(V)]$ or $[T/T_0(p)]$, throughout the state diagram of this phase.[7] At medium and higher pressures, $p\gtrsim 10-12$ atm, and at temperatures $T\lesssim 0.30-0.35$ K, the theory predicts heat-capacity anomalies[8] which have not been investigated experimentally so far. They are seen to fall in the easily accessible He^3 dilution-refrigerator temperature range.

This phenomenological approach can be exploited in connection with the currently unfolding experimental attempts[9-12] toward an initial analysis of a transformation process of liquid He^3 at very low temperatures estimated at $T\lesssim 3$ mK. This extension of the above formalism is tied to the assumption that the transformation is connected with a new ordering of the spin system. It is then expected to be accompanied by an additional reduction in the mean number of excited or free spins. Let $\chi_-(T, V)$ denote the susceptibility in the ordering region $T<T_c(V)$, where $T_c(V)$ is the empirical transition temperature along the indicated isochore. Then, by Eq. (1),

$$\chi_-(T, V)=N_{\sigma-}(T)\mu^2/VkT<\chi_+(T_c, V), \tag{6}$$

χ_+ referring now to $T\geq T_c(V)$, since

$$N_{\sigma-}(T)\leq N_{\sigma+}(T_c), \quad T<T_c\,. \tag{7}$$

At $T \gtrsim T_c$, with $T_c \ll T_0$, χ_+ is essentially χ_p, or by Eq. (4) the susceptibility is constant, independent of the temperature. It exhibits a plateaulike branch at low enough temperatures. On the stated assumption on the nature of the transformation at $T \lesssim T_c$, the susceptibility should fall with decreasing temperatures in the transformation region, being continuous with an angular point at T_c.

By Eq. (5),

$$S_{\sigma-}(T)/Nk = (\ln 2)(N_{\sigma-}/N), \qquad (8)$$

and by Eqs. (7), (1), (2), and (4),

$$\lim_{T\to T_c} \frac{dS_{\sigma-}}{dT} > \left(\frac{dS_{\sigma+}}{dT}\right)_{T_c} = \left(\frac{3}{2}\ln 2\right)\left(\frac{Nk}{T_0}\right). \qquad (9)$$

The assumed spin ordering thus leads to a spin heat-capacity component C_σ such that

$$C_{\sigma-}(T \to T_c) > C_{\sigma+}(T_c) \qquad (10)$$

or to a heat-capacity discontinuity across T_c. If, as assumed, the transformation at T_c were determined by the spin system of the liquid alone, then this discontinuity $[C_{\sigma+}(T_c) - C_{\sigma-}(T_c)]$ or $(\Delta C_\sigma)_{T_c}$ would be finite at least under reduced-temperature resolution around T_c.

At $T \gg T_c$ or $T > T_c$, the heat capacity $C_{n\sigma+}$ arising from the entropy component denoted by us earlier[1,5,8] as $S_{n\sigma}$, referring to thermal disorder other than S_σ, was shown to be only a limited fraction of $C_{\sigma+}$. This suggests the dominant role played by the spin system in the transformation around T_c, and giving rise to the discontinuity $(\Delta C_\sigma)_{T_c}$. This conclusion seems to be in qualitative agreement with some recent preliminary heat-capacity data[10] on compressed liquid He^3. However, the lack of a reliable temperature scale at very low temperatures suggests that this agreement must be considered highly conditional at the present time.

The relation (1) giving the mean fractional number of those thermally excited spins which succeeded in freeing themselves from the interactions within the system is necessarily independent of the strength of the magnetic field used in the measurements of $\chi(T)$, i.e., the susceptibilities $\chi(T, V)$ or $\chi(T, p)$ are independent of the magnetic field. At $T < T_c$, this requires that $\mu H \ll kT_c$. However small, the application of an external, constant and uniform magnetic field $H \ll (kT_c/\mu)$, has the effect to partially polarize magnetically the liquid. This, in turn, is equivalent to depressing the transition temperature $T_c(0)$ in absence of the field. Indeed, on magnetization through raising the field up to the finite if small strength H, the relevant potential energy of ordering is reduced by the energy of magnetization. If $kT_c(H)$ is the approximate potential energy of ordering per atom at $T \lesssim T_c(0)$, at the low field strength H,

$$N_{\sigma-}(T \lesssim T_c)kT_c(H) \simeq N_{\sigma+}(T_c)kT_c(0) - \tfrac{1}{2}V\chi_-(T_c)H^2, \qquad (11)$$

and

$$\lim_{T\to T_c,\ H \ll (kT_c/\mu)} T_c(H) = \left(\frac{N_{\sigma+}(T_c)}{N_{\sigma-}(T_c)}\right)T_c(0) - \frac{\frac{1}{2}V\chi_-(T_c)H^2}{N_{\sigma-}(T_c)k}$$

$$= T_c(0)\left[1 - \left(\frac{H}{H_c}\right)^2\right], \qquad (12)$$

since $N_{\sigma-}(T_c)$ is equal to $N_{\sigma+}(T_c)$ and $V\chi_-(T_c)/N_{\sigma-}(T_c)$ is, by Eq. (6), $[\mu^2/kT_c(0)]$, and we have defined a critical field strength H_c through $[2^{1/2}kT_c(0)/\mu]$. The depression of the ordering temperature $T_c(H)$ parabolic in H at low field strengths is, of course, imposed by the energy of magnetization of the system at these field strengths. Conversely, the very-low-temperature transformation of liquid He^3 at $T < T_c(0)$ should be suppressed at field strengths

$$H(T) > H_c[1 - T/T_c(0)]^{1/2}. \qquad (13)$$

Experimental determination of the magnetic phase-boundary line (12) or (13), near $T_c(0)$, appears accessible through magnetic measurements across $T_c(H)$. At a constant field strength H, the magnetization should fall at $T < T_c(H)$ from a plateaulike branch at $T > T_c(H)$. The locus of the angular points of the magnetization at constant field strengths defines the phase-boundary line.

According to the above formalism of the spin system of liquid He^3, magnetic susceptibility measurements around its apparent transformation point might yield significant information on this new ordering phenomenon.

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[1]L. Goldstein, Phys. Rev. 96, 1455 (1954).
[2]L. Brillouin, J. Phys. Radium 8, 74 (1927).
[3]L. Goldstein and M. Goldstein, J. Chem. Phys. 18, 538 (1950).
[4]L. Goldstein, Ann. Phys. (N.Y.) 14, 77 (1961).
[5]L. Goldstein, Phys. Rev. 112, 1465 (1958); Phys. Rev. 112, 1483 (1958); Phys. Rev. 117, 375 (1960).
[6]L. Goldstein, Phys. Rev. 133, A52 (1965).
[7]The first extensive susceptibility data on liquid He^3 due to A. L. Thomson, H. Meyer, and E. D. Adams [Phys. Rev. 128, 509 (1962)] were shown by us, Ref. 6, to correspond to the predicted universal character of the

susceptibility ratio. For subsequent experimental work see B. T. Beal and J. Hatton, Phys. Rev. 139, A1751 (1965); and H. Ramm, P. Pedroni, J. R. Thompson, and H. Meyer, J. Low Temp. Phys. 2, 539 (1970).

[8]L. Goldstein, Phys. Rev. 148, 108 (1966).

[9]D. D. Osheroff, W. J. Gully, R. C. Richardson, and D. M. Lee, Phys. Rev. Lett. 29, 920 (1972); D. T. Lawson, W. J. Gully, S. Goldstein, R. C. Richardson, and D. M. Lee, Phys. Rev. Lett. 30, 541 (1973).

[10]R. A. Webb, T. L. Greytak, R. T. Johnson, and J. C. Wheatley, Phys. Rev. Lett. 30, 210 (1973); D. N. Paulson, R. T. Johnson, and J. C. Wheatley, Phys. Rev. Lett. 30, 829 (1973).

[11]J. M. Dundon, D. L. Stolfa, and J. C. Goodkind, Phys. Rev. Lett. 30, 843 (1973).

[12]T. A. Alvesalo, Yu. D. Anufriyev, H. K. Collan, O. V. Lounasmaa, and P. Wennerström, Phys. Rev. Lett. 30, 962 (1973); Yu. D. Anufriyev, T. A. Alvesalo, H. K. Collan, N. T. Opheim, and P. Wennerström, Phys. Lett. A 43, 175 (1973).