SPECIFIC HEAT ANOMALY IN SOLID ³He

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A careful look at the data on the specific-heat anomaly in bcc solid 3 He reveals that it disagrees in temperature dependence and concentration dependence with the theory recently proposed by Varma.

Several experiments¹⁻⁴ on solid ³He have indicated that there is an anomalous contribution to the specific heat at temperatures on the order of 0.4° K. Recently Varma⁵ has proposed an explanation of this anomaly. The purpose of this Letter is to review the experimental evidence for the nature of the anomaly and to comment on the explanation due to Varma.

<u>Temperature dependence.</u> – Edwards and Pandorf¹ and Sample and Swenson² have measured the specific heat of bcc ³He at several molar volumes down to $T = 0.30^{\circ}$ K. Both experiments find a quantitatively similar specific-heat anomaly (an excess specific heat) in the temperature range 0.3° K $\leq T \leq 0.5^{\circ}$ K. These experiments measure

$$C(T) = C_p(T) + C_a(T) = \frac{12\pi^4}{5} Nk_{\rm B} \left[\frac{T}{\Theta_{\rm D}(T)}\right]^3, \quad (1)$$

where C(T) is the experimentally observed specific heat, $C_p(T)$ is the phonon specific heat and $C_a(T)$ is the anomalous specific heat. In the temperature range where the anomaly occurs $(T/\Theta_D \le 0.02)$ it is assumed that $C_p(T)$ is proportional to T^3 and that the anomaly must account for any temperature dependence found in $\Theta_D(T)$. From the specific-heat data we have $C_a(0.6^\circ \text{K}) \ll C_p$ (0.6°K) and $C_a(0.3^\circ \text{K}) \approx 0.15 C_p(0.3^\circ \text{K})$.

Adams and co-workers^{3,6} have measured $(dP/dT)_V$ in bcc ³He from above 1°K down to 20 mdeg K. They find a $(dP/dT)_V$ anomaly at temperatures on the order of 0.4°K. More precisely, the experiments of Adams and co-workers measure the excess pressure⁷

$$\Delta P = \Delta P_I + \Delta P_a + \Delta P_p, \tag{2}$$

where ΔP_J is the excess pressure due to the exchange system,

$$\Delta P_I = \operatorname{const} \times \gamma_I J^2 / T, \tag{3}$$

 ΔP_p is the excess pressure due to the phonons, and ΔP_a is the excess pressure due to the anomaly. Panczyk and Adams⁶ have analyzed their data at low temperatures to determine *J*, the constant which characterizes the exchange inter-

action. Their analysis involves extracting the T^{-1} component of the excess pressure at several molar volumes and using Eq. (3). If the excess pressure due to the anomaly were proportional to T^{-1} it would be indistinguishable in its temperature dependence from ΔP_{J} . Then, the data analysis procedure used by Adams and coworkers would determine a $J_{\Delta P}$ which includes ΔP_J and ΔP_a .⁸ The agreement between $J_{\Delta P}$ determined by Adams and co-workers and $J_{\rm NMR}$ determined by NMR experiments strongly suggests that $J_{\Delta P} \approx J_{\rm NMR}$ and that ΔP_a is not large compared with ΔP_J . Perhaps the agreement between $J_{{\Delta}P} \mbox{ and } J_{\rm NMR}$ is fortuitous. In any case ΔP_a proportional to T^{-1} could not lead to an anomaly. It could only lead to a mistaken calculation of $J_{\Delta P}$. But Adams and co-workers find that the anomaly observed at $T \approx 0.4^{\circ}$ K in samples of small molar volume ($V < 22 \text{ cm}^3/\text{mole}$) is much greater than the extrapolation to that temperature of the T^{-1} component of the excess pressure. At larger molar volumes the anomaly persists even after an appreciable correction to the data made by subtracting the extrapolation of the T^{-1} component. Therefore, the excess-pressure anomaly does not depend on temperature as T^{-1} .

<u>Concentration dependence</u>. – The specific-heat anomaly and excess-pressure anomaly are not sensitive to the concentration of ⁴He impurities.

(a) Adams and co-workers³ have looked for ⁴He impurity effects in their excess-pressure data and found none.

(b) The specific-heat data of Edwards and Pandorf¹ at a ⁴He concentration x of 300 ppm compare well with the specific-heat data of Sample and Swenson at $x \approx 2000$ ppm.⁹

(c) The thermal-conductivity experiment of Thomlinson⁴ which gives indirect evidence for the specific-heat anomaly has been performed at x = 2 ppm and x = 100 ppm with no noticeable difference in the results.¹⁰

The theory of the specific-heat anomaly due to Varma disagrees with the experimental observations on two counts; (1) it gives rise to an excess pressure which goes as T^{-1} and (2) it gives rise to an excess pressure (specific heat) which is concentration dependent.

Varma has suggested that an "indirect spin interaction" between pairs of spins due to their coupling to the phonons gives rise to a specific heat contribution of the form

 $C_{\rm is} \sim A^2/T^2$,

where A is a characteristic energy associated with the spin-phonon interaction. The constant A is proportional to J'', the coefficient of the \hat{u}_{ij}^2 term in an expansion of J in terms of the phonon co-ordinates, \hat{u}_{ij} . The J'' term in the expansion of J leads to the "indirect spin-phonon" interaction. This same term is also supposed to be responsible for the two-phonon process which is observed in NMR data at low temperatures.^{11, 12} Its relationship to this data is discussed in detail by Nosanow and Varma.¹³

(1) An excess specific heat proportional to T^{-2} yields an excess pressure proportional to T^{-1} . It is clear from the discussion above that the excess pressure (specific heat) anomaly cannot have this form.

(2) The data of Hatton and Giffard¹² on the "twophonon" relaxation process indicate that it is extremely concentration dependent. In their discussion of these data, Nosanow and Varma¹³ state that this concentration dependence arises from the concentration dependence of J''. Using the Nosanow-Varma analysis of the data of Hatton and Giffard, we must conclude that $(J'')^2$ at x = 2ppm is about 2 orders of magnitude smaller than $(J'')^2$ at x = 200 ppm; $(J'')^2 \propto x$. Since, in the theory of the specific-heat anomaly due to Varma, $C_a(T) \propto (J'')^2$, we have $C_a(T) \propto x$. This prediction is not in agreement with experiment.¹⁴

We believe the theory of the specific-heat anomaly in bcc ³He due to Varma predicts an incorrect temperature dependence and concentration dependence.

The authors wishes to thank Dr. E. D. Adams, Dr. W. J. Mullin, Dr. L. H. Nosanow, and Dr. R. C. Richardson for useful conversations.

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and E. D. Adams, Phys. Rev. Letters <u>23</u>, 518 (1969). ⁴W. C. Thomlinson, Phys. Rev. Letters <u>23</u>, 1330 (1969).

⁵C. M. Varma, Phys. Rev. Letters <u>24</u>, 203 (1970). ⁶M. F. Panczyk and E. D. Adams, Phys. Rev. <u>187</u>, 321 (1969).

⁷The excess pressure and the excess specific heat are simply related. We take the Helmholtz free energy to be $F = k_{\rm B} T f(W/T)$, where W is a characteristic energy. Then $\Delta P = (1/V) \gamma_W CT$, where γ_W is the Grüneisen constant for W.

⁸We use the symbol $J_{\Delta P}$ to mean a value of J calculated from ΔP data; $J_{\rm NMR}$ is a value of J calculated from NMR data.

⁹See also D. O. Edwards, A. S. McWilliams, and J. G. Daunt, Phys. Rev. Letters 9, 195 (1962).

¹⁰In this experiment the relevant temperature range is 0.1° K $\leq T \leq 0.3^{\circ}$ K.

¹¹M. G. Richards, J. Hatton, and R. P. Giffard, Phys. Rev. 139, A91 (1965).

¹²R. P. Giffard and J. Hatton, Phys. Rev. Letters <u>18</u>, 1106 (1967).

¹³C. M. Varma and L. H. Nosanow, Phys. Rev. A <u>1</u>, 133 (1970).

¹⁴Although it is not relevant to the present argument, we wish to point out that the two-phonon part of the data of Richards, Hatton, and Giffard (Ref. 11) and of Giffard and Hatton (Ref. 12) probably has nothing at all to do with a spin-phonon interaction. All features of the data are easily understood in terms of two-phonon scattering from ⁴He impurities. The important point is that the process is not intrinsic to pure ³He.

^{*}Work supported in part by the A. P. Sloan Foundation.

¹R. C. Pandorf and D. O. Edwards, Phys. Rev. <u>169</u>, 222 (1968).