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Adsorbed ^{3}He: Heat Capacity at Low Temperatures*

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WE present here heat-capacity data on adsorbed ^{3}He for different coverages, since they provide general information on the properties of adsorbed helium. These properties are expected to result from a competition between the high zero-point energy of the atoms and their interaction with the adsorbent. From adsorption isotherm data, a number of investigators[1] have concluded that adsorbed helium at low coverages (one layer or less) has a higher density than the bulk liquid, with a solidlike character. This point of view is also supported by interpretation of thermal boundary-resistance measurements between a solid and liquid helium.[2] However, it has been pointed out in a recent paper[3] that the evidence is not conclusive and that adsorbed helium might actually show fluidlike properties. The present data help to clear up this point.

The cryostat has been described elsewhere.[4] The adsorbent, Type 13X zeolite in the form of pellets, $\frac{1}{16}$-in. diameter, was placed in thermal contact with a copper cell through a thin layer of silicone vacuum grease. The cell communicated with a ^{3}He gas-handling system through a stainless-steel capillary that was thermally anchored to the ^{4}He bath and to the cooling salt. The heat capacity of the empty cell was first measured; thereafter, increasing amounts of ^{3}He were admitted to the cell. Corrections for heat exchange with the surroundings were made in the usual way by using the temperature drifts both before and after a heating period.

Figure 1 shows the heat capacity (hc) of different amounts of ^{3}He adsorbed on 0.183 g of zeolite plotted as hc/T versus T, where T is the temperature; the heat capacity of the empty cell has been subtracted. Curves A, B, and C are for 1.9×10^{-4}, 4.7×10^{-4}, and 8.3×10^{-4} mole ^{3}He, respectively.

The high scatter is due to the small capacity of the ^{3}He compared to that of the empty cell. For the smaller coverages A and B, the points lie roughly on a straight line in the hc/T plot, showing a T^2 dependence of the heat capacity. The interaction between the atoms

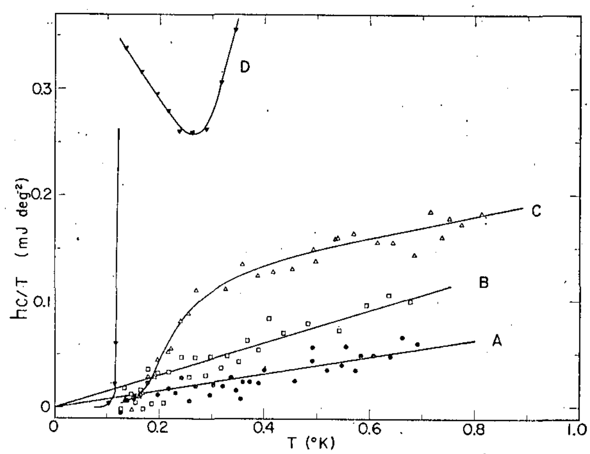

FIG. 1. Heat capacity of He^3 adsorbed on zeolite. For explanation see text.

and the adsorbent is expected to be stronger than the van der Waals forces between He atoms. We can therefore think of a two-dimensional solid with displacements occurring only in the plane of the lattice. The specific heat would then be hc$\simeq$29 $R(T/\theta_D)^2$. According to this formula, our data yield $\theta_D=26$°K compared to $\theta_D=18$°K for the bulk solid at 25 atm. One should, however, note that the zeolite used for this experiment has a rather complicated structure, so that the model used above may not be too realistic. Data for higher coverages (Points D) are different in two respects; (1) hc increases very fast above 0.3°K; no measurements could be made above 0.4°K because the cell cooled down much faster after a heating period than expected from the heat exchange with the surroundings. We interpret this behavior as follows: once the adsorbent is saturated, ^{3}He gas escapes from the cell as the temperature increases and condenses in that part of the capillary in thermal contact with the cooling salt, giving rise to a latent heat contribution to the measured heat capacity. (2) hc presents a sharp peak at 0.12°K which is probably related to isotopic ordering. A similar effect has been observed in the bulk solid[5] as well as in the liquid phase.[6] Our ^{3}He contained 0.04% of ^{4}He, and the ordering temperature observed for this concentration fits very well with other data.[6b] The "bump" shown by Curve C is probably due to the beginning of this process. That no such contribution is observed for the lower coverages indicates that the atoms are strongly bound to their sites. These observations may be compared with recent experiments[7] on superfluid flow in unsaturated ^{4}He films: onset temperatures below 0.7°K have been observed for coverages corresponding to $P/P_0 \sim 10^{-2}$ (P is the pressure at equilibrium of the adsorbed phase; P_0 is the vapor pressure of the bulk liquid at the same temperature). The P/P_0 values for our curves A and B must be of the order of 10^{-4} or less, as no desorption is observed up to about 1°K. No contradiction can therefore be inferred as to the mobility of the adsorbed atoms.

In conclusion, the present data indicate that adsorbed He at low coverages has solidlike properties, in agreement with conclusions drawn from adsorption-isotherm measurements. No nuclear (^{3}He) contribution to the heat capacity is apparent down to 0.1°K.

These experiments have been performed in the laboratory of Professor N. E. Phillips. We wish to thank him for his hospitality during our visit.

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