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NUCLEAR SPIN ORDERING IN ADSORBED He³

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This Letter presents specific-heat data on adsorbed He³ in the temperature range 0.1 to 1.4°K. The results indicate that the nuclear spin entropy is substantially removed at 0.2°K, and emphasize the importance of exchange effects at high densities.

The properties of the bulk solid are not clearly understood. The theory of Bernardes and Primakoff¹ predicts an antiferromagnetic transition around 0.1°K for low pressures (~30 atm) and a ferromagnetic transition at higher pressures (~150 atm), owing to the increased importance of the repulsive term in the interatomic potential. Further calculations by Saunders² yield an exchange energy of about 0.005°K near the melting curve, favoring an antiferromagnetic alignment. Specific-heat³ and susceptibility^{4,5} data show that no magnetic transition occurs at low densities above ~0.02°K; furthermore, no maximum in the melting curve has been observed down to 0.03°K.⁶ At higher densities ($p \sim 100$ atm), susceptibility measurements alone seem to indicate an antiferromagnetic ordering,⁴ with, occasionally, positive deviations from Curie's law.

Adsorption isotherms suggest that helium has a high density in the adsorbed phase,⁷ the interaction with the wall partly overcoming the repulsive action of zero-point motion. The present work was started in the hope that the properties of adsorbed He³ might give some information on the importance of exchange forces at high densities. The adsorbent used was type 13X Zeolite⁸ in the form of pellets 1/16 in. in diameter. Ther-

mal contact with a copper cell was achieved with silicone vacuum grease. The cell was pumped for six hours under high vacuum at 170°C; He³ was then adsorbed at 4.2°K and the cell sealed before introduction in the cryostat. The relative amounts of He³ (34 ± 3 cm³ STP) and adsorbent (0.185 ± 0.005 g) were such as to form approximately a monolayer.

The low-temperature calorimetry technique will be described elsewhere.⁹ The heat capacity of the empty cell was measured separately and has been subtracted. The data are shown in Fig. 1 as a plot of C/T versus T .

The main feature is the sharp peak with a maximum at 0.57°K. The area below this peak is consistent with the removal of the total nuclear spin entropy $S_n = R \ln 2$.

Another maximum is observed at 0.115°K. Relaxation effects were present in this temperature range, the time to attain thermal equilibrium after a heating period being 700 sec at 0.1°K,

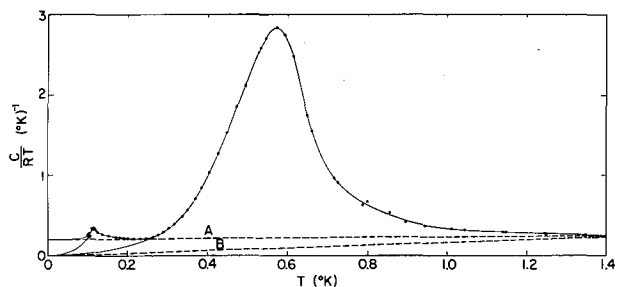


FIG. 1. Specific heat of He³ adsorbed on Zeolite.

400 sec at 0.12°K, and 100 sec at 0.14°K. Accordingly, the points below 0.14°K may be too high as overheating of the cell could not be avoided. These observations are very likely associated with isotopic ordering, as reported by Edwards, McWilliams, and Daunt² in αHe^3 . Our sample had a He^4 concentration $x = 0.21\%$.

The relaxation effects probably arise from a restricted diffusion in the adsorbed phase. (No ordering was observed by Edwards, McWilliams, and Daunt in the β phase which, according to nmr data,¹⁰ has a higher activation energy for diffusion.)

Two straight lines were used (see Fig. 1) to calculate the entropies from the C/T vs T plot, both joining the experimental data at 1.4°K. Line A has a nonzero intercept at $T=0$, and thus assumes a linear term in the specific heat. The area between this line and the full curve yields an entropy $S_n/R = 0.70$, compared to the $\ln 2$ value 0.693. The entropy of mixing similarly obtained from the area below the little peak is $S_i/R = 0.0065$; this is smaller than the calculated value

$$S_i/R = -[x \ln x + (1-x) \ln(1-x)],$$

$$= 0.015 \text{ for } x = 0.0021.$$

However, part of the sample may not take part in the process, the helium atoms being "frozen" by interaction with the walls. If line B is used, and the C/T data extrapolated to the origin, the spin and mixing entropies are, respectively, $S_n/R = 0.82$ and $S_i/R = 0.026$, both higher than the calculated values. Although the above discussion is qualitative, it is likely that a linear term is present in the specific heat, of magnitude $C/RT \sim 0.2$ ($^\circ\text{K}$)⁻¹ compared to the bulk liquid value $C/RT \sim 3$ ($^\circ\text{K}$)⁻¹.

Similar measurements were made on adsorbed He^4 . The specific heat is very small [$C/RT < 10^{-2}$ ($^\circ\text{K}$)⁻¹] below 0.5°K and rises steadily above.

A quantitative interpretation of the present data in terms of exchange integral is difficult, neither the density nor the structure of the adsorbed He^3 being known accurately. As pointed out earlier, the transition is expected to be ferromagnetic at high densities. Nmr experiments have been performed on the same system,¹¹ showing no deviations from Curie's law above 2°K. An extension of these experiments below 1°K would be desirable.

We are currently investigating the transition for different coverages.

These experiments have been performed in the laboratory of Professor N. E. Phillips. We thank him for his encouragement and kind hospitality that made this work possible.

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