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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 Primakoff[1] 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 Saunders[2] yield an exchange energy of about 0.005°K near the melting curve, favoring an antiferromagnetic alignment. Specific-heat[3] 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 Zeolite[8] 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,
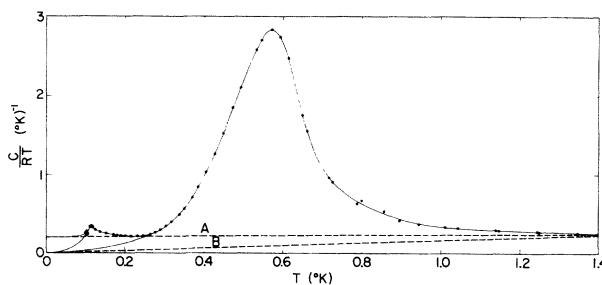

FIG. 1. Specific heat of He³ adsorbed on Zeolite.

400 sec at 0.12°K, and 100 sec at 0.14°K. Ac-
cordingly, 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 Daunt[2] in αHe[3].
Our sample had a He[4] concentration $x = 0.21\%$.

The relaxation effects probably arise from a
restricted diffusion in the absorbed 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 ln2 val-
ue 0.693. The entropy of mixing similarly ob-
tained 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
~ 0.2 (°K)$^{-1}$ compared to the bulk liquid value
$C/RT \sim 3$ (°K)$^{-1}$.

Similar measurements were made on adsorbed
He[4]. The specific heat is very small $[C/RT$
$< 10^{-2}$ (°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 per-
formed on the same system,[11] showing no devia-
tions from Curie's law above 2°K. An extension
of these experiments below 1°K would be desir-
able.

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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