Anomalous Specific Heat of Solid Deuterium
Below 0.6 K*

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An anomaly has been observed in measurements of the specific heat at saturated vapor pressure $C_s$ of pure, solid $D_2$ of low para-$D_2$ concentration below 0.6 K. The new anomaly has been interpreted as evidence for the significance of electric quadrupole–quadrupole interaction between next-nearest-neighbor pairs of para-$D_2$ molecules.

We have extended our previous measurements$^1$ of the specific heat $C_s$ of pure solid $D_2$ with para-$D_2$ concentration of approximately 3.5% to temperatures below 0.6 K. The results are shown in Fig. 1, which plots the specific heat $C_s$ at saturated vapor pressure, against temperature. In Fig. 1 the points denoted by squares give the new data, which extend down to 0.25 K. The new data were taken with a calorimeter system similar to that described previously,$^1$ cooled below 1 K by a $^3$He dilution refrigerator.

In the temperature range 0.6 K–3 K the new results confirm the previous ones$^1$ and can be interpreted in terms of (a) a lattice term $C_{lat} = AT^3$, with $A = 1.32 \text{ mJ/mole-K}^4$ corresponding to a Debye characteristic temperature $\theta_D = 114 \text{ K}$, and (b) an excess specific heat $C_E$ having a broad maximum about 1.4 K.

The excess specific heat $C^E$ is shown in Fig. 2, in which the points marked by squares are the new data. In the temperature range 0.9–5 K, $C^E$ can be fully explained as being due to the electric quadrupole–quadrupole (EQQ) interaction between nearest-neighbor (nn) clusters (pairs and triples) of para-$D_2$ molecules. This interpretation has been fully discussed in our previous paper,$^1$ and the “fixed pairs and triples model” described there leads to an excess specific heat $C^E_I$ as shown in Fig. 2. Clearly in the temperature range 0.9–5 K this model based on EQQ interaction with nn pair and triple clusters reflects the data adequately.

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Below 0.9 K, as is evident from Fig. 2, the experiments cannot be described by the simple model referred to above. The experimental data indicate that below 0.6 K, $C_s$ begins to rise again and $C_s$ must go through a maximum at a temperature below 0.3 K.

To interpret these new lower temperature data, we have extended the EQQ cluster interaction models to include next-nearest-neighbor (nnn) clusters. In the calculation of this extended model the probability of finding a para-D$_2$ molecule as a member of an isolated nearest-neighbor (nn) pair of para-D$_2$ molecules in a hcp lattice turns out to be $P_p = 12x(1 - x)^2$, where $x$ is the para-D$_2$ concentration, as set out by Kreitman and Barnett. This is different from that used in the earlier model, which set $P_p = 12x(1 - x)^{18}$.