EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

NUCLEAR MAGNETIC RESONANCE MEASUREMENTS OF SOLID METHANE DURING THE CONVERSION TO ITS GROUND STATE

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SUMMARY

The conversion of methane to its ground state at 1.06°K was observed by NMR. The signal intensity and spin-lattice relaxation time indicate that a slow conversion to the ground state J = 0, I = 2 occurs.

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In previous studies on solid methane at liquid helium temperature, there was some doubt whether the molecular rotational and spin quantum numbers J and I were good quantum numbers in the solid[1-6]. If this is the case, the ground state should be $J = 0$ and $I = 2$. The $I = 2$ state is of special interest for the purpose of using methane as a polarized target substance[7], as its thermal equilibrium polarization is twice as high as that of free protons, in the high temperature approximation.

In order to solve this question, we compared directly the absolute value of the RF susceptibility of the solidified methane with that of other compounds containing hydrogen. The measurements were made at 1.06°K in a magnetic field of 25 kG. The NMR signal was found to increase during about 50 hours to an equilibrium value which was indeed twice as big as that expected from the free proton density. In order to obtain some information about the nature of this slow signal increase, the proton spin-lattice relaxation time T_1 was measured at short intervals. T_1 is of the order of 1 sec, so that the slow signal increase with time cannot be attributed to spin-lattice relaxation. Typical results are shown in the diagram. T_1 has a characteristic maximum about 40 minutes after cooling the sample and a minimum 1 hour later, followed by an increase to an equilibrium value during two days. Although this general behaviour was observed in every sample, details of this variation seemed to depend on several parameters, such as the way of cooling the sample, and on the sample holder used, and they could not be reproduced. The samples contained about 1‰ oxygen; a sample containing about 1% oxygen reached equilibrium in about two hours, with a final value of $T_1 = 0.5$ sec.

Our measurements of the increase in the RF susceptibility are in agreement with previous results[1,2]. De Wit[3,4] found a much shorter T_1 in the oxygen-free methane used by him. This dependence of T_1 on oxygen impurity agrees with theoretical predictions[5] that the high-temperature spin species distribution is frozen in in the oxygen-free methane. In that case, the strongly relaxing species I = 1 has the heavy statistical weight of 9/16. This explanation is also supported by the results of Hopkins et al.[2], who found no significant signal increase in oxygen-free methane.

No important increase was observed in samples containing about 15% air. These samples had a line-width of 30 gauss, compared to the line width of about 6 gauss in the normal samples. This indicates that the molecular rotation is hindered if some air is mixed with the methane. This possibility of obtaining different kinds of frozen methane according to the degree of purity might explain some contradictory results from previous studies[8-10].

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Figure Caption

Fig. 1 : Signal intensity and spin-lattice relaxation time of protons in solid methane as a function of time after freezing.