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NUCLEAR SPIN-LATTICE RELAXATION AND EVIDENCE FOR A LOW TEMPERATURE PHASE TRANSITION IN SOLID $n$-D$_2$

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Abstract.- We present new results for the nuclear spin-lattice relaxation times in solid $n$-D$_2$ at low temperatures. There is an abrupt change in the temperature dependence of the relaxation times at $T \approx 170$ mK and we interpret this in terms of a possible transition to a quadrupolar glass phase.

Recent low temperature N.M.R. experiments on solid H$_2$ at low ortho hydrogen concentrations /1-5/ have shown evidence for the existence of transitions to a quadrupolar glass phase in which the orientational degrees of freedom of the ortho molecules are frozen at random. We present here preliminary results of a study of the nuclear spin-lattice relaxation times of solid n-D$_2$($X(J=1)=0.33$) which show an abrupt change in their temperature dependence analogous to the changes observed in solid H$_2$ for low ortho H$_2$ concentrations /1/.

The interest in solid H$_2$ and solid D$_2$ lies in their quantum crystal aspects. The orbital angular momentum $J$ is a good quantum number and at low temperatures only the lowest $J$ values $J=0$ and $J=1$ need to be considered. Spin statistics require that we consider two molecular species: ortho H$_2$($J=1$, I=1) and para H$_2$($J=0$, I=0) and para D$_2$($J=1$, I=1) and ortho D$_2$($J=0$, I=0 and I=2). I$ is the total nuclear spin. At high temperatures the $J=1$ molecules behave as an assembly of weakly interacting rotators but for temperatures $T<3K$, the anisotropic intermolecular interactions (principally electrostatic quadrupole-quadrupole) result in a collective orientational ordering of the $J=1$ molecules. The EQQ interaction of an isolated pair is minimized for a "tee" configuration but it is impossible to realize a 3-D close packed lattice with all the molecules mutually perpendicular. The system is "frustrated" /6/ and for pure ortho H$_2$ and pure para D$_2$ the total free energy is minimized when the molecules are aligned parallel to the body diagonal $\mathbf{e}_a$ of an f.c.c. lattice /7/ (Pm$_3$ space group). The ordered state is characterized by the order parameters $\langle 3J^2 \rangle_{Z}$ and one must use many body quantum crystal techniques in order to account for the zero point fluctuations of the orientations of the molecules.

As the concentration $X$ of the $J=1$ molecules is reduced, the zero point fluctuations increase /8/, the frustration decreases and the features of the long range orientationally ordered Pa$_3$ phase are no longer observed below a critical concentration, $X_c(H_2)=0.55$ /9/ and $X_c(D_2)=0.59$ /10/. For concentrations $X<X_c$, N.M.R. experiments on solid H$_2$/2, 4/ have shown that as the temperature is lowered, there is a transition to a quadrupolar glass phase in which the orientational degrees of freedom are frozen locally. The N.M.R. spectra can be interpreted in terms of a broad distribution of local order parameters $\sigma_i=\langle 3J^2 \rangle_{Z_i}$ where the local axes $\mathbf{e}_i$ vary at random from one site to another. The spin 1 quadrupolar glass phase observed in solid H$_2$ provides an interesting challenge for the spin glass theories.

In order to obtain further experimental evidence and additional information on the quadrupolar glass phases we sought a second example of this phase and we have carried out N.M.R. studies on solid n-D$_2$. In the ordered phase of solid D$_2$ at high para concentrations one observes an important narrow central line (2 kHz width) due to the $I=2(J=0)$ spins superimposed on a weaker broad line (75 kHz

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width) attributed to the $I=1$ spins ($J=1$) /1/ C.W. studies of solid D$_2$ for $X = 0.33$ show no evidence of the anticipated broad $I=1$ spectra at low temperatures ($60<T<400$ mK) and for this reason we tried to detect possible phase transitions in solid n-D$_2$ by studying the relaxation times.

The relaxation of the $J=1$ species is due to the modulation of the nuclear dipole and quadrupole interactions resulting from fluctuations of the molecular orientations. In the disordered phase the relaxation times $\tau_{1L}/13/ \propto \exp(\Delta/\lambda)$. The phase transitions are thus accompanied by abrupt changes in the temperature dependence of the $\tau_{1L}$. The orientational degrees of freedom are in the also frozen quadrupolar glass phases at low $X$ and dramatic changes in the temperature dependence of the relaxation time in solid H$_2$ are observed at the transition temperature /2/.

In solid D$_2$ the $I=2(J=0)$ spins relax much more slowly than the $I=1(J=1)$ spins /12/ and the observed relaxation of the $I=2$ spins in the disordered phase /13/ has been interpreted in terms of a cross-relaxation ($\tau_{12}$) with the $I=1$ spins followed by the spin-lattice coupling of the latter. An abrupt change in the temperature dependence of the $\tau_{1L}$ at the transition should then be observable by studying the temperature dependence of the relaxation of the central part of the N.M.R. spectrum.

We have studied the relaxation times of solid n-D$_2$ for $60<T<500$ mK at 25 MHz. A typical recovery from partial saturation is shown in figure 1 where one can identify two distinct relaxation rates $\tau_s$ (short) and $\tau_L$ (long). At high temperatures $\tau_{1L}<\tau_{2L}<\tau_{12}$, the 2-bath model predicts $\tau_s = \tau_{1L}$ and $\tau_L \propto \tau_{12}/\mu$ where $\mu = \tau_{12}/\tau_{21} < 5$. At low temperatures one expects that $\tau_{12} < \tau_{1L} < \tau_{2L}$ and therefore $\tau_s \propto \tau_{12}$ and $\tau_L \propto \tau_{1L}/\mu$. The $\tau_s$ and $\tau_L$ obtained from these recoveries for several temperatures are shown in figure 2 and are consistent with this interpretation. There is a dramatic change in the temperature dependence of the relaxation rates at $T = 170$ mK similar to that observed in solid H$_2$ /1/.

These results provide indirect evidence for a possible transition to an ordered phase for $X=0.33$. We plan to extend these studies to higher para concentrations and in particular study the low temperature N.M.R. lineshapes near the critical concentration.
References