

# NMR Studies of the Dynamics of Orientational Glasses<sup>1</sup>

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## I. Introduction

There are a number of simple well-defined molecular systems for which the centers of mass form a periodic lattice structure, but for which the orientational degrees of freedom of the molecular units appear to be "frozen" at random with no long-range correlations (1). These **orientational glasses** include solid ortho-para H<sub>2</sub> mixtures (2), N<sub>2</sub>-Ar mixtures (3) and KBr<sub>1-x</sub>K(CN)<sub>x</sub> mixed crystals (4), and they continue to generate intense interest because of the apparently **universal low-temperature properties** observed for a very diverse range of examples. Many of the properties observed bear strong resemblance to those seen for the general family of spin glasses (5), (dilute magnetic alloys, mixed crystals [Eu<sub>x</sub>Sr<sub>1-x</sub>S (6), Rb<sub>1-x</sub>(NH<sub>4</sub>)<sub>x</sub>H<sub>2</sub>PO<sub>4</sub> (7), partially doped semiconductors (8), Josephson junction arrays (9), and others).

The most striking universal features are an apparent freezing of the local degrees of freedom on

long time scales without any average periodic long-range order, characteristic slow relaxations, history dependence following external field perturbations (magnetic, electric, elastic-strain, etc.) and a very large number of stable low energy states (which increase monotonically with the number of active atomic or molecular centers). The existence of a true thermodynamic phase transition has been questioned, but a number of observations of the canonical spin glasses (such as CuMn) point to a thermodynamic freezing of the spin degrees of freedom and the transition is not purely kinetic. Detailed studies of the dynamics in these systems is crucial for developing a better understanding of the fundamental physics of these glassy systems, and the orientational glasses are particularly interesting as test cases because the dynamics can be probed in detail by techniques such as nuclear magnetic resonance.

In this review we will develop a unified quantitative treatment of the low temperature thermal behavior and the explicit time dependence of the low-frequency relaxations in the orientational glass state of solid hydrogen. This was the first orientational glass to be discovered and serves as a prototype for the other orientational systems.

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## II. Orientational Order Parameters

The order parameters needed to specify the degrees of freedom of interacting quantum rotors such as ortho-H<sub>2</sub> are more complex than those needed to describe the canonical spin glasses (10,11). For the spin glasses we consider only the dipole moments  $\langle S_x \rangle_i, \langle S_y \rangle_i, \langle S_z \rangle_i$  of the spin  $S$  at the lattice site  $i$ . For the quantum rotors with angular momentum  $J = 1$ , we need to specify the quadrupolar moments  $\langle J_\alpha J_\beta \rangle_i$  in addition to the vectorial components  $\langle J_\alpha \rangle_i$ . Only five of the quadrupole moments are independent variables;  $\alpha$  and  $\beta$  represent one of the Cartesian components ( $x, y, z$ ) of the local reference frame.

In the absence of interactions that break time-reversal symmetry, the orbital angular momentum is quenched in the molecular solids, *i.e.* the  $\langle J_\alpha \rangle_i$  vanish for all  $\alpha$ . In order to determine the five parameters needed to fully describe the molecular degrees of freedom, we therefore select as local molecular reference axes ( $x, y, z$ ) the principal axes of the quadrupolar tensor

$$Q_{\alpha\beta} = \left\langle \frac{1}{2} J^2 \delta_{\alpha\beta} - \frac{3}{4} (J_\alpha J_\beta) \right\rangle \quad (1)$$

In this principal axes frame,  $Q_{xy} = Q_{yz} = Q_{zx} = 0$ , and the only intrinsic quadrupolar order parameters are (12)

(i) the **alignment**

$$\sigma_i = Q_{zz} = \left\langle 1 - \frac{3}{2} J_z^2 \right\rangle_i \quad (2)$$

and (ii) the **eccentricity**

$$\eta_i = \langle J_x^2 - J_y^2 \rangle_i \quad (3)$$

The five order parameters are the three local reference axes (giving the **direction** of the molecular orientations) and the two quadrupolar parameters  $\sigma$  and  $\eta$  which give the **degree** of alignment with respect to those axes.

There are restrictions on the allowed values of  $\sigma$  and  $\eta$ . If the single particle density matrices are expressed in terms of  $\sigma$  and  $\eta$ , the strong positivity condition that the eigenvalues of the density matrix be positive-definite, leads to the conditions

$$\frac{1}{3} + \frac{2}{3}\sigma \geq 0 \quad (4)$$

$$\frac{1}{3} - \frac{1}{3}\sigma \pm \frac{2}{3}\eta \geq 0$$

This is discussed in detail in references (11) and (13).

All of the experimental data has been analyzed assuming local axial symmetry, *i.e.*, that  $\eta = 0$ . This is not justified on theoretical grounds and the assumption is made for simplicity only. Experimental tests for nonvanishing  $\eta$  have been proposed (11), but they have not been carried out.

The NMR methods used by several groups to study ortho-para-H<sub>2</sub> mixtures, N<sub>2</sub>/Ar mixtures and related systems, can provide direct measurements of the local order parameters. The CW absorption spectra give the NMR second moments  $M_2(T)$  which provide the equivalent of the Edwards-Anderson glass order parameter, and the nuclear spin-lattice relaxation rates  $T_1^{-1}$  and spin-spin relaxation rates  $T_2^{-1}$  provide information about the autocorrelation functions  $\langle\langle \sigma_i(t) \sigma_i(0) \rangle\rangle_{av}$ . These assertions result from the fact that the intramolecular nuclear dipole-dipole interaction  $H_{DD}$  is directly related to the order parameters. If the orientational degrees of freedom are expressed in terms of irreducible tensorial operators  $T_{LM}$  that transform analogously to the spherical harmonics  $Y_{LM}$  in the manifold  $J = 1$ , and if the nuclear spin degrees of freedom are expressed in terms of corresponding tensorial operators  $N_{LM}$  in the manifold  $I = 1$ ,

$$H_{DDi} = D \sum_M T_{2M_i} N_{2M_i}^+ \quad (5)$$

where  $D = 173.1$  kHz,

$$T_{20} = \frac{1}{\sqrt{6}} (3J_z^2 - J^2), \quad (6)$$

$$T_{21} = \frac{1}{2} (J_z J_+ + J_+ J_z), \text{ and}$$

$$T_{22} = \frac{1}{2} J_+^2$$

$$T_{LM} = (-)^M T_{L,-M}^+ \text{ and}$$

$$N_{20} = \frac{1}{\sqrt{6}} (3I_z^2 - I^2), \text{ etc.}$$

If the molecules are orientationally disordered,  $H_{DD}$  averages to zero, and only a single NMR line is observed at the Larmor frequency. If the molecule has a preferred orientation,  $H_{DD}$  does not average to

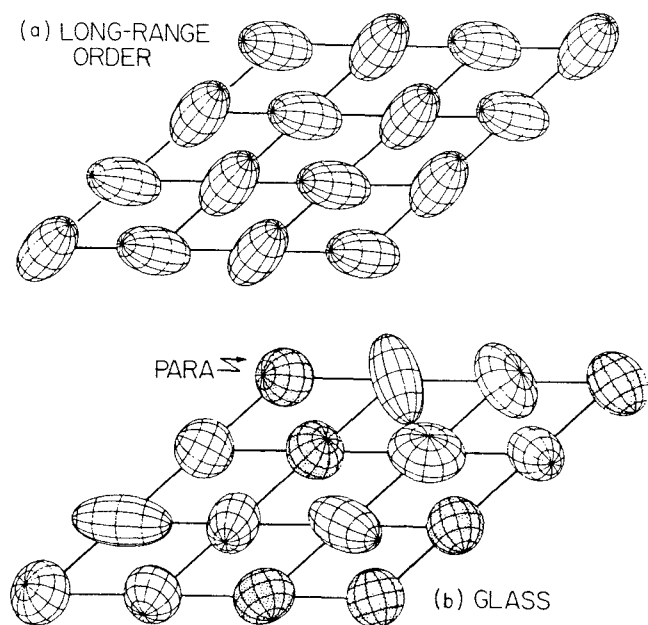


Figure 1: Schematic representation of orientational degrees of freedom in (a) the quadrupolar glass phase of solid hydrogen for ortho contents less than 55%, and (b) the long range ordered  $Pa_3$  structure for concentrations above 55%.

zero, and each molecule contributes a doublet to the NMR spectrum with components at frequencies

$$\Delta\nu_i = \mp \frac{3}{2} D \langle T_{20}(i) \rangle_Z \quad (7)$$

The expectation value is evaluated with respect to the magnetic field axis  $Z$  and **not** the local symmetry axis  $z$  with respect to which the molecular order parameters are defined. Effecting the transformation to the local molecular frame  $(x, y, z)$ , we find

$$\Delta\nu_i = \mp D [-\sigma_i P_2(\cos \theta_i) + \frac{3}{4} \eta_i \sin 2\theta_i \cos 2\phi_i] \quad (8)$$

$(\sigma_i, \phi_i)$  are the polar angles defining the orientation of the applied magnetic field with respect to the local molecular symmetry axes. With the assumption of local axial symmetry ( $\eta_i = 0$ ) and a powder distribution for the angles  $(\theta_i, \phi_i)$ , the NMR spectra can be interpreted in terms of a broad distribution of local order parameters  $P(\sigma)$  for the local alignments  $\sigma$ .

A schematic representation of the orientational glass phase deduced from the NMR observations for solid ortho-para  $H_2$  mixtures is shown in Figure 1. The prolate ellipsoids represent molecules

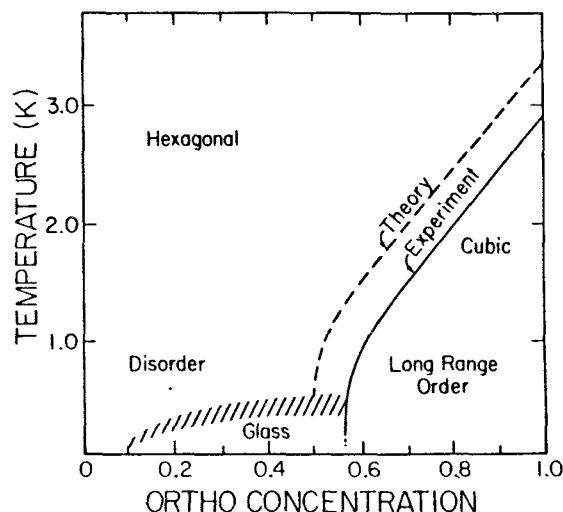


Figure 2: Phase diagram for the observed orientational ordering in solid ortho-para  $H_2$  mixtures as a function of ortho concentration. The hatched region designates the smooth transition from the glass phase to the rotationally disordered phase.

which are aligned preferentially along local axes, while the oblate ellipsoids represent molecules that are aligned preferentially perpendicular to the local axes  $O_z$ . It is important to note that **both** the molecular axes  $(x, y, z)$  and the intrinsic quadrupolar order parameters  $\sigma_i$  vary at random throughout the sample. From this point of view, dilute ortho-para- $H_2$  mixtures form a true **quadrupolar glass** rather than an orientational glass. For the latter, only the local axes  $(x, y, z)$  would be distributed at random.

The quadrupolar glass phase is only observed for ortho- $H_2$  concentrations less than 55% (Figure 2). Above 55%, one observes long range orientational order in an antiferro-orientationally ordered  $Pa_3$  structure with first order transitions to the high temperature rotationally disordered phase. The transitions from the glass phase to the disordered phase are smooth with no indication of an abrupt phase transition, but the orientational degrees of freedom appear to become frozen over a relatively small temperature range shown by the hatched region of Figure 2.

The absence of a distinct phase transition from the glassy state to the rotationally disordered phase has raised a number of questions concerning the nature of the quadrupolar glass system in solid ortho-para  $H_2$  mixtures. Is the "freezing" of the orien-

tational degrees of freedom a cooperative process similar to that which is believed to occur for the canonical spin glasses, or is it simply a local thermodynamic evolution determined by fixed local fields which vary throughout the sample? It is important to note that these quenched solid-molecular systems do not display broken symmetry. Solid H<sub>2</sub> remains hcp down to the lowest temperature studied for ortho concentrations less than 55%, and the random occupation of lattice sites leads to the existence of local electric field gradients, the field conjugate to the local order parameter, and this field plays the same role as the magnetic field for the dipolar spin glasses. This random local field therefore makes the problem of local orientational ordering in random mixtures equivalent to the local dipolar ordering in spin glasses in the presence of random magnetic fields, and thus any phase transition would be smeared out. The answer to these questions has been provided by a systematic study of the low frequency dynamics of the glass phase which has provided a unified quantitative explanation of the low temperature behavior of the heat capacities and the explicit time dependence of the glass-like relaxation at low temperatures in terms of the low energy density of states for the system.

### III. Stimulated Echo Studies of Low Frequency Dynamics

Nuclear-spin-stimulated echoes have proved to be very useful for the study of ultraslow molecular motions both in molecular orientational glasses (14) and in the ordinary ("window pane") glasses (15). The stimulated echoes are prepared by a three-pulse sequence. Two initial rf pulses separated by a short time  $\tau$  prepare the system with a "fingerprint" of the local order parameters  $\sigma_i$ . The local nuclear-spin temperatures following a  $90_y - \tau - 90_y$  sequence depend on  $\sigma_i$  and vary as  $\cos(D_i\sigma_i\tau)$  where  $D_i = DP_2(\cos\theta_i)$ . The detailed calculations are given in reference (11). The single particle nuclear spin density matrix just after the first pulse is

$$\rho_i(t = 0^+) = 1 + \frac{\hbar\omega_L}{k_B T} I_{x_i} \quad (9)$$

where  $\omega_L$  is the nuclear Larmor frequency. Only the term involving  $I_x$  contributes to the NMR responses and in the following we consider only the evolution

of the off-diagonal component

$$\rho'_i(t = 0^+) = \frac{\hbar\omega_L}{k_B T} I_{x_i} = -\frac{\hbar\omega_L}{k_B T} (N_{1,1} - N_{1,-1})_i \quad (10)$$

where we have used the irreducible tensorial operators of eqn. 6 to obtain the last expression. After the first pulse, the evolution of the density matrix under the influence of the intramolecular dipolar Hamiltonian  $H_{DD}$  leads to the production of both vectorial and second rank tensorial components of the nuclear magnetization

$$\rho'_i(t = \tau) = -\frac{\hbar\omega_L}{k_B T} [N_{1,1}(\tau) - N_{1,-1}(\tau)]_i \quad (11)$$

where

$$N_{1,\pm 1}(\tau) = \cos(D_i\sigma_i\tau)N_{1,\pm 1} \\ \mp j \sin(D_i\sigma_i\tau)N_{0,\pm 1}$$

The second pulse applied at time  $\tau$  is used to convert the transverse components  $N_{L,\pm 1}$  into longitudinal components  $N_{L,0}$ . If the "waiting" period  $t_w$ , before applying the third pulse is long compared to the transverse relaxation time  $T_2$ , but short compared to the nuclear spin-lattice relaxation time  $T_1$ , all transverse components  $N_{L,\pm 1}$  die away before the application of the third pulse and one is left with **only** the secular components  $N_{L,0}$ . If we consider the general case of a second pulse which rotates the spins through an angle  $\Psi$  about an axis in the  $x - y$  plane making an angle  $\Phi$  with the  $y$ -axis, the secular component of the density matrix after the second rf pulse is

$$\rho'_{io}(t = \tau^+) = -\sqrt{2} \frac{\hbar\omega_L}{k_B T} [\cos\Phi \cos(D_i\sigma_i\tau) \times \\ \sin\Psi N_{1,0,i} - \sqrt{\frac{3}{4}} \sin\Phi \sin(D_i\sigma_i\tau) \sin^2\Psi N_{2,0,i}] \quad (12)$$

The important point is that the *amplitude* of the  $N_{L,0,i}$  depends on the *initial values* of the local order parameters through the terms  $D_i\sigma_i\tau$  established during the short preparation period  $\tau$ . The nuclear spin polarizations  $N_{1,0}$  and nuclear spin alignments  $N_{2,0}$  therefore carry a "fingerprint" of the initial values of the local order parameters. This fingerprint is interrogated at a later time  $t_w$  by the third rf pulse which converts the stored secular components into transverse components  $N_{L,\pm 1}$ . These components then evolve under the intramolecular Hamiltonian at that later time. If the order parameters have not

changed during  $t_w$ , the evolution during a time  $\tau$  after  $t_w$  will match that during the preparation time, and the spin components will focus to form a stimulated echo at time  $t = T_w + 2\tau$ . If the parameters change, however, the echo is attenuated, and this damping of the echo amplitude is used to follow the changes in the order parameters during the waiting period.

The stimulated echo formed from the secular components of the density matrix is called the primary stimulated echo. If nonsecular components remain during  $t_w$ , i.e. if  $t_w \leq T_2$ , additional echoes can be formed and these are called secondary stimulated echoes (11).

If the order parameters during the preparation period are designated by  $\sigma_i(0)$  and the values of the parameters at time  $t_w$  by  $\sigma_i(t_w)$ , the amplitudes of the stimulated echoes for different pulse sequences are given by:

$$(a) 90_y - \tau - 90_y - t_w - 90_y - \tau$$

$$S_x = -\frac{\hbar\omega_L}{k_B T} \langle\langle \cos[D_i\sigma_i(0)\tau] \cos[D_i\sigma_i(t_w)\tau] \rangle\rangle \quad (13)$$

$$S_y = 0$$

$$(b) 90_y - \tau - 45_x - t_w - 45_x - \tau$$

$$S_x = \frac{3}{4} \frac{\hbar\omega_L}{k_B T} \langle\langle \sin[D_i\sigma_i(0)\tau] \sin[D_i\sigma_i(t_w)\tau] \rangle\rangle \quad (14)$$

$$S_y = 0$$

$$(c) 90_y - \tau - 90_y - t_w - 90_x - \tau$$

$$S_x = 0$$

$$S_y = -\frac{\hbar\omega_L}{k_B T} \langle\langle \cos[D_i\sigma_i(0)\tau] \cos[D_i\sigma_i(t_w)\tau] \rangle\rangle \quad (15)$$

$$(d) 90_y - \tau - 45_x - t_w - 45_y - \tau$$

$$S_x = 0$$

$$S_y = -\frac{3}{4} \frac{\hbar\omega_L}{k_B T} \langle\langle \sin[D_i\sigma_i(0)\tau] \sin[D_i\sigma_i(t_w)\tau] \rangle\rangle \quad (16)$$

where  $S_x$  and  $S_y$  refer to the in-phase signal and quadrature signals, with respect to the phase of the initial rf pulse. The double brackets refer to an ensemble average for all the molecules.

The most striking result observed for the decay of the stimulated echoes has been a characteristic logarithmic decay seen at long times for low temperatures (Figure 3). This behavior has been observed

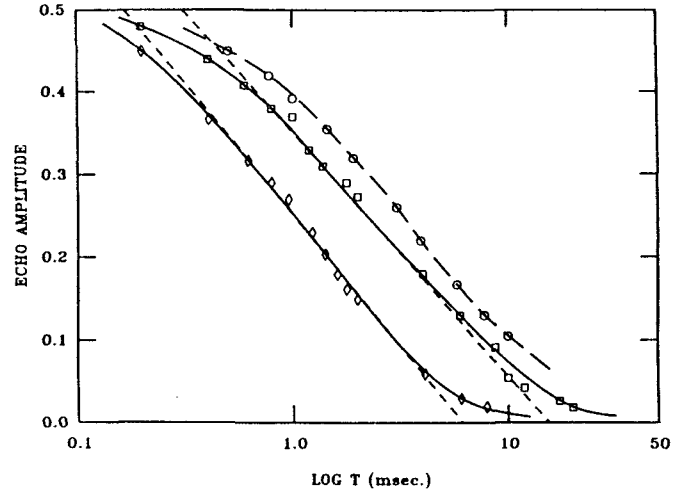


Figure 3: Observed long-time decay of stimulated echoes in the orientational glass phase of solid ortho-para  $H_2$  mixtures. squares,  $D\tau = 1.7$ ; diamonds  $D\tau = 3.5$  (Ref.[14],  $T = 0.22$  K  $X_{ortho} = 0.54$ ); circles  $D\tau = 7.77$  (Ref.[16],  $T = 0.153$  K,  $X_{ortho} = 0.43$ ). The dashed lines refer to the calculated decays using the heat capacity data, Ref. (17).

for both solid ortho-para  $H_2$  mixtures and for solid  $N_2/Ar$  mixtures and is believed to be a characteristic signature of the slow "glassy" relaxation of the molecular orientations in these systems. The results can be understood in terms of a model of collective low energy excitations which appear to be universal to the orientational glasses. This interpretation is consistent with thermodynamic measurements carried out on the same systems and is discussed in the next section.

## IV. Low Energy Excitations

The characteristic logarithmic decay of the stimulated echo amplitudes at long times is shown in Figure 3 for solid ortho-para  $H_2$  mixtures. This logarithmic decay can be understood quantitatively in terms of a scaling model introduced by Fisher and Huse (18) for short-range spin glasses. In this model, the long-distance and long-time correlations at low temperatures are dominated by long-time excitations associated with the coherent reorientation of clusters of correlated molecules.

Consider a small connected cluster of  $N$  molecules which are reoriented with respect to a reference ground state (which is possibly nonunique). A connected excitation consists of a single wall enclosing

a connected cluster. We are interested in the low-frequency or long length-scale excitations which involve the coherent collective reorientations of a large number of connected molecules. The basic assumption of Fisher and Huse (18) that the density of excited states for a cluster of size  $L$  scales as  $L^{-\theta}$  with  $0 \leq \theta \leq \frac{(d-1)}{2}$  allows one to consider the thermally excited clusters as dilute, and therefore regarded as "non-interacting two level systems". If the free energy barriers for the formation of the clusters is  $E_B$ , the tunneling rate for a characteristic cluster at temperature  $T$  is

$$\Gamma(E_B) = \Gamma_0 \exp\left(-\frac{E_B}{k_B T}\right) \quad (17)$$

where  $\Gamma_0$  is the characteristic attempt frequency.

In the long-time limit,  $\Gamma_0$  is reasonably well defined because it is associated with a characteristic cluster dimension. The dynamics of the clusters must be hierarchical because the low energy barriers (for small clusters) must be overcome before the larger ones can be attempted. In this model, therefore, the dynamics associated with the faster degrees of freedom constrain the slower degrees of freedom. In a time  $t$ , the only barriers crossed will be those for which  $0 < E_B < E_{max}(t)$ , where from eqn. 17

$$E_{max}(t) = k_B T \ln(\Gamma_0 t) \quad (18)$$

For all barriers crossed in time  $t$  there will be significant changes in the local order parameters, and for the molecules in the clusters participating in the excitation, the average correlation functions

$$\langle\langle \cos[D_i \sigma_i(0)\tau] \cos[D_i \sigma_i(t_w)\tau] \rangle\rangle$$

will vanish. The contribution to the stimulated echoes from these molecules will therefore also vanish, while the contribution from molecules with larger energy barriers will still contribute to the echo. Summing over the density of low energy excitations,  $P(E_B)$ , we therefore find for the echo amplitude at time  $t$

$$S_x(t) = \left(\frac{\hbar\omega_L}{2k_B T}\right) \left[1 - \int_0^{E_{max}(t)} P(E_B) dE_B\right] \quad (19)$$

At very low temperatures, we need only consider the density of states near  $E = 0$ . The density of states can be determined from an analysis of the heat capacity measurements at low temperatures.

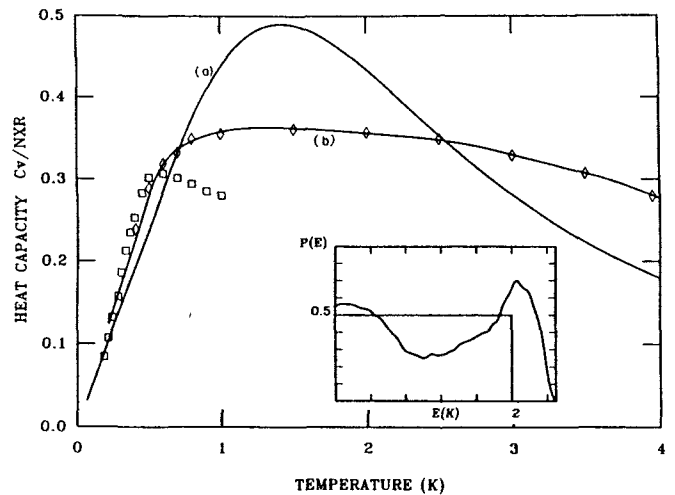


Figure 4: Observed heat capacities in the orientational glass phase of solid  $H_2$ . [diamonds Ref.(19,20); squares Ref. (21) line (a) is for a flat distribution  $P(E)$  of energy levels, and line (b) is for a  $P(E)$  adjusted to fit the high temperature data. The inset shows the two corresponding distributions for  $P(E)$ .

As in almost all glass-like systems,  $P(E)$  is approximately constant at low energy, and the echo amplitude at time  $t$  is therefore

$$S_x(t) = \frac{\hbar\omega_L}{k_B T [1 - k_B T P(0) \ln(t/T_0)]} \quad (20)$$

where  $T_0 = (\pi/D\tau\Gamma_0)$ . The slopes of logarithmic decays shown in Figure 3 are therefore directly proportional to the density of low energy excitations. The observed decay shown in Figure 3 for an ortho fraction  $\dot{x} = 0.54$  at 0.22 K corresponds to a density of states  $P(0) = 0.59 \pm 0.05 \text{ K}^{-1}$

In order to test this interpretation, we have compared the density of states  $P(0)$  inferred from the stimulated echo decay with that deduced from heat capacity measurements in the orientational glass phase of solid hydrogen. The experimental capacities are shown in Figure 4. Lines (a) and (b) in Figure 4 are the calculated heat capacities assuming a simple two level system in which each ortho- $H_2$  molecules of angular momentum has energy states  $J_{\zeta_i} = \pm 1$  separated from the state  $J_{\zeta_i} = 0$  by an energy gap  $\Delta_i$ . ( $\zeta_i$  is the local axis of symmetry of the molecules for which  $|\sigma_i|$  is maximum.) Line (a) is for a hypothetical flat distribution,  $P(E) = \text{constant}$ , and line (b) is a numerical fit to the experimental data. The density of states at  $E \simeq 0$  deduced from the heat capacity data is  $P(0) = 0.55 \pm 0.08 \text{ K}^{-1}$

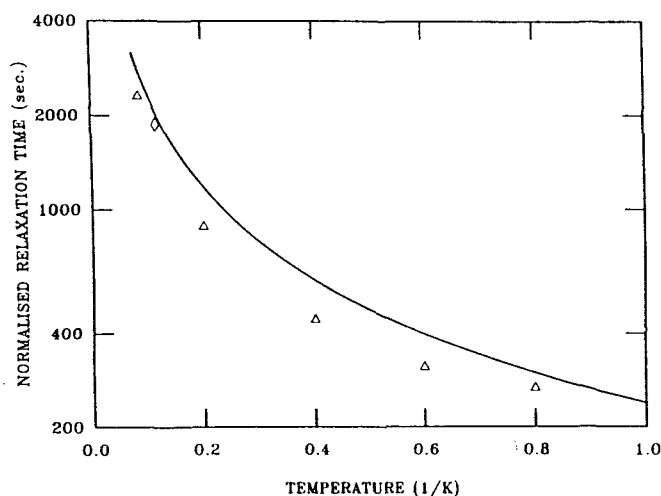


Figure 5: Temperature dependence of the nuclear spin-lattice relaxation rate in the orientational glass phase of solid  $H_2$ . [triangle, Ref. (22); diamond, Ref.(16)]

and this is in excellent agreement with the value  $P(0) = 0.59 \pm 0.05 \text{ K}^{-1}$  inferred from the NMR data. The consistency of the analysis of the NMR results with the thermodynamic data confirms the view that the total density of excited states at low energies is dominated by the large-scale excitations.

An additional test of the model is provided by the observed temperature dependence of the nuclear spin-lattice relaxation times in the glass phase. The most effective relaxation process at very low temperatures is a Raman process involving the simultaneous excitation and de-excitation from two excited states which differ by the nuclear Zeeman energy. This relaxation can be calculated using the density of states estimated above from the thermodynamic analysis and the NMR results, and the comparison with the observed rates is shown in Figure 5. The calculations are in good qualitative agreement with the measurements, and it should be noted that other relaxation mechanisms that have been proposed have failed to describe the temperature dependencies even semi-quantitatively (22).

## V. Conclusion

The excellent agreement between the predicted NMR stimulated echo decays and the observed variation shows that the low frequency dynamics in the orientational glass state of solid  $H_2$  can be described in terms of low-energy large-scale cluster ex-

citations. The available density of states for these excitations is approximately constant at very low temperatures and is consistent with thermodynamic measurements. It will be important to test this model by carrying out systematic studies of related glass-like systems where the freezing of the degrees of freedom is also determined by frustrated short-range interactions in random mixtures.

## VI. Acknowledgments

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