Quadrupolar glass freezing in solid hydrogen: Distribution functions for the orientational order parameters

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The microscopic model for quadrupolar glass freezing of axial quadrupoles in solid hydrogen is considered in the mean-field approximation. While the glass order parameter exhibits continuous growth with decreasing temperature and the quadrupolar susceptibility a smeared-out behavior, stability analysis indicates a well-defined nonergodic region of a quadrupolar glass phase with many degenerate thermodynamic states with irreversibility phenomena. We also analyze effects due to quantum-mechanical tunneling between different quadrupole orientations at low temperatures. Furthermore, by employing self-consistent equations for the order parameter we obtain the distribution function P(p) for the local quadrupolarizations p which describe the extent to which the molecular orientations of ortho species are frozen. The temperature evolution of P(p) shows a continuous growth of the local order parameter p with a broad shape of the distribution function and with no evidence for a sharp phase transition.

I. INTRODUCTION

There has been considerable experimental effort during the past decade to understand the orientational ordering in dilute molecular systems such as solid ortho-para hydrogen mixtures.¹ These systems represent random arrays of interacting quadrupoles, which provide striking examples of disorder and frustration. In the case of the solid-H₂ mixtures only the ortho-H₂ molecular species with orbital angular momentum J = 1 are orientable. The para species with J = 0 are spherically symmetric, thus have no electric quadrupole moment and play the role as an almost perfect dilutant. In order to characterize the orientational order an order parameter has to be defined. In the case of solid hydrogen the lowest multipole moment of the molecular orbital is the quadrupole moment being a tensorial quantity. Of five linearly independent quadrupolar components, three can be used to define local axes, thus two nonvanishing quadrupole parameters $p(T) = \langle 3J_z^2 - 2 \rangle_T$ and $\eta(T) = \langle J_x^2 - J_y^2 \rangle_T$ represent intrinsic degrees of freedom, with J_{α} ($\alpha = x, y, z$) being the orbital angular momentum operators and $\langle \cdots \rangle_T$ the statistical average, respectively. Here, p(T) measures the alignment along the z axis, while $\eta(T)$ is a measure for the departure from axial symmetry and is called eccentricity. At high ortho concentrations the ordered state is characterized by a long-range order parameter p. However, for ortho-hydrogen concentrations less than approximately 55%, nuclear magnetic resonance (NMR) studies and thermodynamic measurements showed no evidence of orientational ordering for temperatures T > 0.4 K. Instead, NMR experiments at very low temperatures show broad spectra whose features have been interpreted in terms of the freezing of the orientational degrees of freedom of the ortho molecules into a state called guadrupolar glass.² Information on the local quadrupolar order parameters are obtained from NMR data, making use of the fact that NMR line shape is determined by the intramolecular dipole-dipole interaction between the nuclear spins.³ This interaction depends directly on p and η and on the polar angles between the local axes and the magnetic field. The shape of the NMR profile is related to the distribution of the local order parameters P(p). Neglecting the eccentricity one has

$$P(p) = \frac{1}{N} \sum_{i} \delta(p - \langle 3J_z^2 - 2\rangle_T)$$
$$= [\delta(p - \langle 3J_z^2 - 2\rangle)]_{av}$$
(1.1)

where $[\cdots]_{av}$ denotes the average over the disorder present in the system. Equation (1.1) implies that P(p) is self-averaging, i.e., the two types of averaging in Eq. (1.1) are equivalent.

In analogy to magnetic (dipolar) spin glasses one can introduce the Edwards-Anderson⁴ (EA) order parameter for quadrupolar systems as

$$q_{\rm EA}(T) = [\langle 3J_z^2 - 2\rangle 2_T]_{\rm av} .$$
 (1.2)

This quadrupolar glass parameter also can be determined directly from the NMR data as it is directly given by the second moment of P(p),

$$q_{\rm EA} = \int dp p^2 P(p). \tag{1.3}$$

The distribution P as a function of both quadrupole parameters p and η was derived by Li, Meyer, and Berlinsky⁵ based on an approximation to the one-particle orientational density matrix for the randomly distributed J = 1 molecules. However, this theory still remains

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phenomenological since the properties of the distribution function depends on the single temperature-dependent fit parameter.

In the present paper we take a different approach in that we start from the microscopic model for the quadrupolar glass and derive the shape of P(p) within the full self-consistent theory. For simplicity we restrict ourselves to the axial case. In Sec. II we introduce the microscopic Hamiltonian for the quadrupolar glass in terms of the infinite-range Sherrington-Kirkpatrick⁶ model for randomly interacting quadrupoles. Subsequently, we obtain self-consistent equations for glass order parameters. We show that within the mean-field approach the equations for orientational glass order parameter have a nonzero solution in the whole temperature and/or field range, i.e., do not indicate phase transition. However, the smooth behavior of the order parameter does not exclude the possibility of having well-defined quadrupolar glass transition, which marks the appearance of many degenerate thermodynamic states separated by infinitely high barriers. A similar situation occurs, for example, in magnetic spin glasses in the presence of external fields or nonzero mean of the bond probability distribution.⁷ Experimentally, such glass features manifest as strong metastability and irreversibility in response to external fields. Therefore, another objective of ours is to determine the validity of the mean-field approach by performing the stability analysis in Sec. III. Finally, in Sec. IV, we present predictions for the distribution functions of local quadrupolarizations resulting from the microscopic theory.

II. HAMILTONIAN AND DISORDER AVERAGE

We consider in the present paper, in the spirit of the Sherrington-Kirkpatrick⁶ model, the infinite-range version of the uniaxial quadrupolar system⁸ with the Hamiltonian

$$H = -\sum_{i,j} J_{ij} Q_{zi} Q_{zj} - \Omega \sum_{i} J_{xi}$$
(2.1)

where $Q_{zi} = 3J_{zi}^2 - 2$ and J_{zi}, J_{xi} denote quadrupolar and orbital angular momentum J = 1 operators, respectively, at lattice site *i*. Furthermore, the last term in Eq. (2.1) describes the quantum-mechanical tunneling between different quadrupole orientation, with Ω being the tunneling frequency, in analogy to quantum dipolar case.⁹ The latter effect might be relevant at very low temperatures where the quantum nature of the system becomes apparent. The J_{ij} are quenched, independently distributed exchange interactions with the probability distribution

$$P_J(J_{ij}) = (N/2\pi J^2)^{1/2} \exp(-NJ_{ij}^2/2J^2)$$
(2.2)

with N being the number of lattice sites. It should be pointed out that the randomness of the quadrupolar interaction between two ortho-H₂ molecules is not of the same type as the randomness resulting from Rudderman-Kittel-Kasuya-Yosida (RKKY) interaction in the conventional (magnetic) spin-glass problem.¹⁰ However, since the orientations of the ortho-H₂ molecule at a given site relative to the local coordinate system (chosen in a way to coincide with the principal axes of the quadrupolar momentum tensor) varies randomly from site to site, the coupling constant J_{ij} itself becomes a random quantity. In the limiting situation of long-range order in a nearly pure J = 1 solid this picture will change, since molecular alignments would be along well-defined directions. Fortunately, for ortho-para hydrogen mixtures at ortho-H₂ concentrations, relevant for quadrupolar-glass features, it is plausible to suppose that the distribution for the couplings J_{ij} will be Gaussian about zero mean. The assumption that the interactions between the molecules are infinite ranged is, of course, a first-order approximation. A more sophisticated theory would require us to include the wave-vector dependence as well as the influence of spatial fluctuations. However, within the theoretical framework for spin glasses it would be extremely difficult to incorporate short-range interaction effects even for the simplest Ising-like models. The virtue of the assumption about infinite-interaction range allows us to proceed along the canonical procedures developed for the conventional spin glasses. 6

Following the standard route of the thermo field dynamic (TFD) approach for the quantum spin-glass problem,¹¹ we shall discuss the thermodynamics of the system in terms of the disorder averaged generating functional for the TFD causal Green's functions¹² in functional representation

$$\langle Z[\eta, \{J_{ij}\}] \rangle_J = \int \prod_{ab} DR^{ab} \exp(-NL[\mathbf{R}] + \Lambda[\eta])$$
(2.3)

where $Z[\eta, \{J_{ij}\}]$ is the unaveraged generating functional for fixed realization of random bonds and $\Lambda[\eta] =$ $\text{Tr}(\mathbf{R}\eta)/J^2$ denotes the source term. Specifically in the interaction picture with respect to the single-body Hamiltonian $H_0 = -\Omega \sum_i J_{xi}$ one obtains the effective action $L[\mathbf{R}]$ in the form

$$L[\mathbf{R}] = \mathrm{Tr}\mathbf{R}^2 - \ln\Phi[\mathbf{R}] \tag{2.4}$$

where

$$\operatorname{Tr} \mathbf{R}^{2} = \int_{-\infty}^{+\infty} dt \int_{-\infty}^{+\infty} dt' \sum_{ab} R^{ab}(t,t') R^{ba}(t',t), \qquad (2.5)$$

while $\Phi[\mathbf{R}] = \langle 0, \beta \mid U_R(-\infty; +\infty) \mid 0, \beta \rangle$ with $\langle 0, \beta \mid \dots \mid 0, \beta \rangle$ = Tr exp $(-\beta H_0 \dots)/\text{Tr} \exp(-\beta H_0), \beta = 1/kT$, and the time-ordered exponential reads

$$U_R(-\infty; +\infty) = T_t \exp\left(-i \int_{-\infty}^{+\infty} dt \int_{-\infty}^{+\infty} dt' H_R(t, t')\right)$$
(2.6)

with the effective Hamiltonian

$$\hat{H}_{R}(t,t') = -\sum_{ab} (\epsilon_{a}\epsilon_{b})^{1/2} J R^{ab}(t,t') Q_{z}^{a}(t) Q_{z}^{b}(t'). \quad (2.7)$$

Here, a, b = 1, 2 are the TFD "dynamic replicas" ($\epsilon_1 = 1, \epsilon_2 = -1$) (Ref. 11) labeling the collective fields R^{ab}

which act as dynamic self-interactions in the effective Hamiltonian (2.7) between time-dependent quadrupolar operators $Q_z^a(t) = \exp(-iH_0t)Q_z^a \exp(iH_0t)$.

As usual in the $N \to \infty$ limit the steepest descent method can be used which amounts in finding the stationary point R_0^{ab} determined by the extremal conditions $\delta L[\mathbf{R}]/\delta R^{ab} = 0$. Thus, one obtains a self-consistent equation for the collective variable $R_0^{ab}(t, t')$,

$$R_0^{ab}(t,t') = \frac{1}{2} (\epsilon_a \epsilon_b)^{1/2} J g^{ab}(t,t')$$
(2.8)

where

$$g^{ab}(t,t') = -i \frac{\langle 0,\beta \mid TQ_{z}^{a}(t)Q_{z}^{b}(t')U_{R_{0}}(-\infty;+\infty) \mid 0,\beta \rangle}{\langle 0,\beta \mid TU_{R_{0}}(-\infty;+\infty) \mid 0,\beta \rangle}.$$
(2.9)

The onset of the glassy phase is marked by a nonzero value of the spin-glass order parameter. Within the context of the dynamic theory the spin-glass order parameter has to be determined via time-persistent quantities. Therefore, we factorize the matrix of the causal TFD Green's functions into finite-time $(g_{reg})^{ab}$ and time-persistent parts $(g_{sing})^{ab}$ as follows: $g^{ab}(t,t') = (g_{reg})^{ab}(t-t') + (g_{sing})^{ab}$, and the correspondence with physical observables is achieved by the following decomposition of the Fourier transformed causal Green's function in the space of thermo-field components (cf. Ref. 11),

$$(g_{\rm reg})^{ab}(\omega) = [\tau \overline{g}(\omega)]^{ab} - \frac{2i(C_{\rm reg})(\omega)}{e^{\beta\omega} + 1} \begin{pmatrix} 1 & e^{\beta\omega/2} \\ e^{\beta\omega/2} & 1 \end{pmatrix}$$
(2.10)

while

$$\overline{g}^{ab}(\omega) = \begin{pmatrix} g^{R}(\omega) & 0\\ 0 & g^{A}(\omega) \end{pmatrix}, \qquad \tau = \begin{pmatrix} 1 & 0\\ 0 & -1 \end{pmatrix}$$
(2.11)

with $g^{R(A)}(\omega)$ being the retarded (advanced) Green's functions. Correspondingly, $(C_{reg})(\omega)$ refers to the ther-

modynamic correlation function being related to $g^{R}(\omega)$ by means of the fluctuation-dissipation theorem. Furthermore, it turns out that the time-persistent part $(g_{\text{sing}})^{ab}(\omega)$ has the form

$$(g_{\rm sing})^{ab}(\omega) = -2\pi i q \delta(\omega) \begin{pmatrix} 1 & 1\\ 1 & 1 \end{pmatrix}^{ab} .$$
 (2.12)

Combining Eq. (2.10) and (2.12) one obtains

$$C(\omega) = (C_{reg})(\omega) + 2\pi q \delta(\omega)$$
(2.13)

in accordance with the standard dynamic definition of the EA spin-glass order parameter.⁴

III. ORDER PARAMETERS

The time-persistent contribution to the effective Hamiltonian can be represented by using auxiliary Gaussian integration having the form of a static random field to generate time-persistent autocorrelation. Accordingly, from the condition (2.9) one obtains the selfconsistent equations for the spin-glass order parameter qand the quadrupolar susceptibility χ ,

$$q = \langle p^2(x) \rangle, \quad \chi = \langle \chi(x) \rangle , \qquad (3.1)$$

where $\chi = \lim_{\omega \to 0} \langle g_R(\omega, x) \rangle$ is the static quadrupolar susceptibility, $g_R(\omega, x)$ is the unaveraged dynamic response function, and the Gaussian average over the static noise is given by

$$\langle \cdots \rangle = \int_{-\infty}^{\infty} \frac{dx}{(2\pi)^{1/2}} \exp(-x^2/2) \dots$$
 (3.2)

The analytical expressions for the x-noise-dependent quadrupolarization p(x) and susceptibility $\chi(x)$ in the general case $(kT \neq 0, \Omega \neq 0)$, while available, are too massive to be presented here. Therefore, we consider the classical and quantum limits separately. For the clasical case $(\Omega = 0)$ one obtains correspondingly

$$p(x) = 2 \frac{\exp\left(\frac{3Jq^{1/2}x}{kT}\right) - \exp\left(\frac{3J^2\chi}{2kT}\right)}{2\exp\left(\frac{3Jq^{1/2}x}{kT}\right) + \exp\left(\frac{3J^2\chi}{2kT}\right)}$$
(3.3)

and

$$\chi(x) = \frac{1}{kT} \frac{18 \exp\left(\frac{3\Theta_x}{2kT}\right)}{4 \exp\left(\frac{6Jq^{1/2}x}{kT}\right) + 4 \exp\left(\frac{3\Theta_x}{2kT}\right) + \exp\left(\frac{3J^2\chi}{kT}\right)}$$
(3.4)

while

$$\Theta_x = 2Jq^{1/2}x + J^2\chi \ . \tag{3.5}$$

In the pure zero-temperature quantum case $(\Omega \neq 0)$ the transition is controlled exclusively by the transverse field and the resulting equations are

$$q = \langle p_0^2(x) \rangle, \quad \chi = \langle \chi_0(x) \rangle \tag{3.6}$$

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with

$$p_0(x) = \frac{1}{2} \left[\frac{18Jq^{1/2}x - 9J^2\chi}{(36J^2qx^2 - 36J^3\chi q^{1/2}x + 9J^4\chi^2 + 16\Omega^2)^{1/2}} - 1 \right]$$
(3.7)

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(4.1)

and

$$\chi_0(x) = \frac{144\Omega^2}{(36J^2qx^2 - 36J^3\chi q^{1/2}x + 9J^4\chi^2 + 16\Omega^2)^{3/2}}.$$

and (3.6). However, in contrast to the dipolar glasses, quadrupolar glass order parameter q exhibit nonzero value in the whole temperature range. Quite interesting behavior arises also in the quantum case ($\Omega \neq 0$) (Fig. 2). Surprisingly, the action of the transverse field Ω does not

suppress quadrupolar glass order parameter q at any fi-

nite value of Ω . This is in contrast to the case of, e.g., the quantum Ising model in a transverse field, where tunneling effects destroy glass order at some critical value of the

transverse field.⁹ We ascribe these behaviors to the non-

vanishing quadrupolar moment which enters Eqs. (3.1)

IV. STABILITY ANALYSIS

In order to locate the transition to the nonergodic

phase we perform the stability analysis by observing that

where $g_R(\omega) = \langle g_R(\omega, x) \rangle$, diverges in the static limit

 $(\omega \to 0)$ at the instability point. From Eq. (2.9) it follows

the generalized damping function $\gamma(\omega)$,

 $\gamma^{-1}(\omega) = i \frac{\partial}{\partial \omega} g_R^{-1}(\omega)$

and (3.6) via χ and couples to quadrupolar operators.

that the dynamic-response function $g_R(\omega)$ in turn obeys a Dyson equation

$$g_R(\omega, x) = \Sigma_R(\omega, x) \left[1 - J^2 g_R(\omega) \Sigma_R(\omega, x) \right]^{-1}$$
(4.2)

Specifically, differentiation of Eq. (4.2) with respect to the frequency ω and subsequent averaging over the static noise results in the relation

$$\begin{split} \left[1 - J^2 \langle g_R^2(\omega, x) \rangle \right] \frac{\partial g_R(\omega)}{\partial \omega} \\ &= \langle \Sigma_R^{-2}(\omega, x) \frac{\partial \Sigma_R(\omega, x)}{\partial \omega} g_R^2(\omega, x) \rangle \quad (4.3) \end{split}$$

which leads to the marginal stability condition in the form

$$1 - J^2 \langle \chi^2(x) \rangle > 0. \tag{4.4}$$

Numerical solution of the self-consistent equations (3.1) and (4.4) together with stability constraint (4.4)reveals critical value $kT_c/J \approx 1.38$ for temperature, below which unstable nonergodic phase is present. A similar calculation for the quantum case [Eq. (3.6)] with the stability condition $1-J^2\langle\chi^2_0(x)
angle>0$ results in the stability threshold for the transverse field $\Omega_c/J \approx 1.53$. In physical terms it means that below these values one enters the nonergodic region (i.e., a multiple-valley structure of the phase space for $N \to \infty$). Precise theoretical description in this region would presumably require a Parisi-like approach of the broken replica symmetry.¹³ Unfortunately, the Parisi scheme successfully applied for the Ising and vector models seems to fail for quadrupolar glass system.¹⁴

1.75 1.5 $q^{1/2}$ 1.25 1 0.75 $J\chi$ 0.5 0.25 n 2 4 6 8 Ω/J

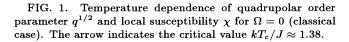
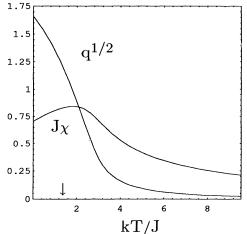
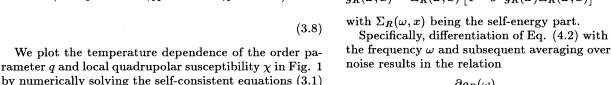
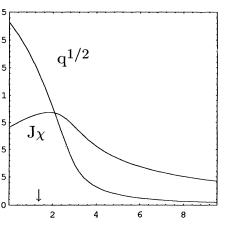


FIG. 2. Transverse-field dependence of quadrupolar order parameter $q^{1/2}$ and local susceptibility χ in the T = 0 case. Indicated by arrow is the critical point $\Omega_c/J \approx 1.53$.







V. DISTRIBUTIONS OF LOCAL QUADRUPOLARIZATIONS

A natural way to analyze the NMR data for ortho-para hydrogen mixtures is to invoke a distribution of order parameters [Eq. (1.1)] to account for the distribution of local quadrupolarizations in the quenched alloy. In the following, we present an evaluation of the distribution of local quadrupolarizations based on the self-consistent equations for the order parameters derived in Sec. III.

In terms of static noise-dependent quadrupolarization p(x) the distribution function for the local quadrupolarization (1.1) becomes

$$P(p) = \int_{-\infty}^{+\infty} \frac{dx}{2\pi} e^{-x^2/2} \delta[p - p(x)] .$$
 (5.1)

In general P(p) satisfies the sum rule which follows from Eq. (5.1),

$$\int_{-2}^{1} dp P(p) = 1 , \qquad (5.2)$$

$$P(p) = \frac{kT}{J} \frac{\exp\left\{-\frac{1}{2q}\left(\frac{kT}{J}\right)^2 \ln^2\left[\frac{\exp\left(\frac{J^2\chi}{2kT}\right)(p+2)^{1/3}}{2^{1/3}(1-p)^{1/3}}\right]}{(2\pi q)^{1/2}(1-p)(p+2)}\right\}}$$

where q and χ are self-consistent solutions of Eq. (3.1). The result of a numerical evaluation of the probability distribution of local quadrupolarizations is presented in Fig. 3 where P(p) is plotted against p in the interval [-2, 1] of the allowed values¹⁵ of quadrupolarization for several temperatures. The temperature evolution of P(p)exhibits a smooth displacement of the distribution center of gravity to lower values of |p| as T increases, with a broad shape for intermediate values of the temperature in qualitative agreement with experiment. For $T \to \infty$ while the second moment is just the quadrupolar glass order parameter

$$q = \int_{-2}^{1} dp p^2 P(p) . \qquad (5.3)$$

For example, the distribution function for the NMR lineshape $G(\nu)$ can be written in terms of the Pake line-shape function $F(\nu, p)$ as follows:¹

$$G(\nu) = \int_{-2}^{1} dp P(p) F(\nu, p) . \qquad (5.4)$$

By using the well-known property of the Dirac delta function

$$\delta[f(x)] = \delta(x - x_0) / |f'(x_0)| \tag{5.5}$$

with x_0 being the solution of the equation f(x) = 0, we can integrate out the static noise component in Eq. (1.1) to give

distribution function approaches $\delta(p)$ whereas for $T \to 0$ P(p) becomes sharply peaked at p = -2 and p = 1.

In order to simulate the effect of quantum effects at very low temperatures we have calculated the distribution function $P_0(p)$ at T = 0 as a function of the transverse field Ω representing the frequency of the tunneling between different quadrupolar states. By using the equation for $p_0(p)$ and taking into account Eq. (5.1) one obtains for the local polarization distribution at zero temperature the following result:

$$P_{0}(p) = \frac{3\Omega}{2J} \frac{\exp\left\{-\frac{\left[2\Omega|2p+1|+3J^{2}\chi(1-p)^{1/2}(p+2)^{1/2}\right]^{2}}{72J^{2}q(1-p)(p+2)}\right\}}{(2\pi q)^{1/2}(1-p)^{3/2}(p+2)^{3/2}} .$$
(5.7)

The function $P_0(p)$ plotted in Fig. 4 has a symmetric shape, which becomes narrower for increasing values of Ω . The broad shape of $P_0(p)$ for finite values of Ω indicates that due to the quantum-mechanical effect at zero temperature perfect alignment at the ground-state configuration of local quadrupolarizations is absent.

VI. SUMMARY

We have considered a microscopic model for orthopara hydrogen quadrupolar glass with axial symmetry. Using the mean-field approach, the self-consistent equations for the quadrupolar glass order parameter and local quadrupolar susceptibility have been derived. Since both parameters do not exhibit any singularity, indicating well-defined glass transition temperature, stability analysis was employed to single-out the nonergodic region. The behavior in quadrupolar glass appears to be somewhat similar to the conventional dipolar glasses in an applied longitudinal field which breaks spin-reversal symmetry. In the later case the sharp transition is also smeared out and the nonergodic region is located below Almeida-Thouless line.⁷ In the quadrupolar glass case, the spin reversal symmetry is broken even in the hightemperature phase without applied field.¹⁶ Referring to experimental work in quadrupolar systems, some exper-

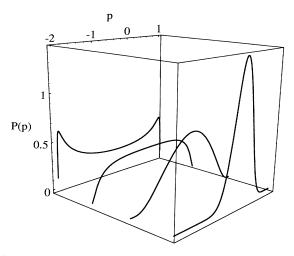


FIG. 3. Distribution function of local quadrupolarizations P(p) for several values of the temperature: kT/J = 1.6, 2, 2.4, 4 (from the left to the right).

imentally available data seems to support the instability picture presented here. For example, observations in the solid ortho-para hydrogen mixtures show substantial slowing down of orientational fluctuations accompanied by hysteretic effects—signaling nonergodic behavior.¹⁷ Since the bulk of the experimental data on ortho-para hydrogen mixtures comes from NMR line shapes, the distribution function of local quadrupolarizations (describing the extent to which the molecular orientations are frozen), which can be extracted from the NMR spectrum, is of special interest. As calculated in the present paper, distributions evolve only smoothly with temperature, which suggests a continuous growth of quadrupolarizations on lowering the temperature giving no hint for an abrupt phase transition. We also analyzed the model at zero temperature with a transverse field to account for the quantum effects. In contrast to previous phenomenological approaches, we performed calculations

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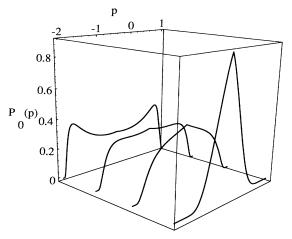


FIG. 4. Distribution function of local quadrupolarizations at zero temperature $P_0(p)$, for different values of the transverse field Ω : $\Omega/J = 1.6, 2, 2.4, 4$ (from the left to the right).

within a self-consistent theory starting from the microscopic model. Despite its relative simplicity, the essential qualitative features of the distribution functions seem to be reproduced. Since there is no reason for the requirement of axial symmetry to be completely satisfied, a more sophisticated treatment should also include the effect of eccentricity, at the cost of introducing the second quadrupolar order parameter. We hope to elaborate on this point in a future work.

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