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Microscopic approach to the quadrupolar-glass problem

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We report results of the microscopic approach to the quadrupolar-glass problem in mixed molecular crystals formed by linear and spherical molecules. The temperature-concentration analysis of the orientational order parameter derived from NMR spectroscopy data is given on the two-component vector model of site-diluted ortho-para-hydrogen-type mixtures. Two features of the short-range orientational ordering, characteristically different from the spin-glass case, namely, incomplete freezing and residual ordering, respectively, at low and high temperatures are discussed. It is established that the first effect is due to spatial correlated local molecular and reaction fields and the second caused by quadrupolar intrinsic field. A zero-field freezing temperature is estimated taking into account the threshold concentration for quadrupolar glass formation.

Ortho-hydrogen (or para-deuterium) molecules possess a unit angular momentum, $J=1$, which remains a good quantum number even in solid state and thus can be treated as quantum rotators or pseudospins.[1] In ortho-para hydrogen mixtures (o-$p\mathrm{H}_2$) spherical symmetric para-hydrogen molecules play a role of almost perfect dilutants[2] and provide a striking effect of random substitution. Molecular hydrogens (H_2, D_2, and HD) being the simplest of all molecular solids give a rich testing ground for the investigations of random substitution and frustration effects on cooperative phenomena in different materials.[3,4] The work of Sullivan *et al.*[5] stimulated active experimental,[3,4,6] theoretical,[3,6–10] and Monte Carlo simulation[11,12] investigations of orientationally disordered hydrogens in the temperature region of short-range-order frozen states originally named by the quadrupolar glass (QG). The QG orientational order parameter, as a direct analog of the Edwards-Anderson spin-glass (SG) magnetic order parameter,[13] can be derived from the second moment of the NMR absorption signal line shape.[3] Its temperature behavior indicates some characteristic features, which make it possible to distinguish between two types of glasses. First, at low temperatures the QG order parameter, observed in a wide concentration region, being extrapolated to zero temperature is far from its maximum value equal to unity,[14,15] which is assumable[16] due to zero-point motion of linear quadrupoles. On the other hand, such a kind of unsaturated orientational order effect of the same order in magnitude also occurs in Ar-N_2 mixtures[17,18] and thus is specific to both quantum and classical QG's. Secondly, even in the absence of external fields the orientational order parameter is different from zero at high enough temperatures.[14] This fact points to the existence of some intrinsic field conjugate to the local parameter order[3] and characteristic of QG's. The main object of this paper is to draw a fundamental distinction between quadrupolar orientational and dipolar spin glasses related to the short-range-order freezing of suitable degrees of freedom.

The pseudospin reformulation of intermolecular interaction originally termed in tensorial quadrupolar dynamic variables[19] has been given for classical[20] and quantum[21] QG systems. In the latter case the QG Hamiltonian for the $(J=1)_c(J=0)_{1-c}$ system can be presented in the following form:

$$\mathscr{H}=-\sum_{f>f'} c_f c_{f'} \sum_{\mu,\mu'=-2}^{2} J_{ff'}^{\mu\mu'} S_{\mu f} S_{\mu' f'} - \sum_{f} c_f \sum_{\mu} H_{\mu f}^{(\mathrm{Cr})} \mathrm{S}_{\mu \mathrm{f}}. \tag{1a}$$

The orientational degrees of freedom of a given quadrupole $(J=1)$ are described by the set of five spherical components of the quadrupolar moment tensor given in the local spherical coordinate system, quantization axes of which coincide with the principal-axis frame of the quadrupolar moment tensor (for details see Refs. 20–26). Thermal expectation values of the dynamic variables are the two-component QG local order parameter:[3,27] $\langle S_{\mu f}\rangle_T=\delta_{\mu 0}\sigma_f+\delta_{\mu 2}\eta_f$. An application of microscopic statistical consideration to distinguish between random-bond and random-site effects — both are due to random-substitutional dilution—makes it possible to present the exchange interaction $J_{ff'}^{\mu\mu'}$ and crystalline field $H_{\mu f}^{(Cr)}$ given in (1) in explicit form [see, respectively, Eqs (6) and (7) in Ref. 21 and Appendix in Ref. 22]. Their nonzero expectation values are

$$\langle J_{ff'}^{\mu\mu'} J_{ff''}^{\nu\nu'}\rangle_B=\Gamma^2\delta_{f'f''}\delta_{\mu\mu'}\delta_{\nu\nu'};$$

$$\langle H_{\mu f}^{(\mathrm{Cr})}\, H_{\mu' f'}^{(\mathrm{Cr})}\rangle_B = h_{\mathrm{Cr}}^2\delta_{ff'}\delta_{\mu\mu'}, \tag{1b}$$

where Γ is electrostatic quadrupole-quadrupole (EQQ) nearest-neighbor coupling constant.[28] The random-bond average, labeled by B, includes the uniform integration over random local-axis directions and the hcp-lattice summation over intermolecular directions, i.e. $\langle\cdots\rangle_B=\langle\langle\cdots\rangle_L\rangle_R$. The configurational average, labeled by C, in turn additionally includes the average over the random-site variables ($\langle c_f\rangle_s=c, \langle c_f c_{f'}\rangle_s=c\,\delta_{ff'}+c^2(1-\delta_{ff'})$, c is concentration of quadrupoles), i.e., $\langle\cdots\rangle_C=\langle\langle\cdots\rangle_B\rangle_S$. In the framework of the two-component ($\mu=0,2\rightarrow\mu,\sigma$) vector model the local order parameters are[21]

$$\sigma_f = 1 - 3\frac{\cosh[(\sqrt{3}/2)\varepsilon_{\eta f}/T]}{2\cosh[(\sqrt{3}/2)\varepsilon_{\eta f}/T] + \exp(\frac{3}{2}\varepsilon_{\sigma f}/T)},$$

$$\eta_f = \frac{\sqrt{3}\sinh[(\sqrt{3}/2)\varepsilon_{\eta f}/T]}{2\cosh[(\sqrt{3}/2)\varepsilon_{\eta f}/T] + \exp(\frac{3}{2}\varepsilon_{\sigma f}/T)}. \tag{2}$$

The configurational average can be performed by means of the effective two-component vector local field ϵ_f, which in turn can be estimated within various mean-field (MF) type approximations. We will calculate it on the base of the MF-type Hamiltonian

$$\mathscr{H}_{\mathrm{MF}} = -\sum_f c_f \sum_{\mu=\sigma,\eta} (\epsilon_{\mu f} + H_{\mu f}^{(\mathrm{Cr})}) S_{\mu f} + 2h_{\mu f}(S_{\mu f} - \sigma_{\mu f})^2, \tag{3a}$$

following from (1) taking into account thermal and spatial fluctuations through the trial molecular $\epsilon_{\mu f}$ and fluctuation $h_{\mu f}$ fields. The desired effective molecular field conjugate to the local order parameters $\sigma_{\mu f}$ (2) can be represented in the form[22]

$$\varepsilon_{\mu f} = \epsilon_{\mu f} + \pi_{\mu f} + H_{\mu f}, \quad \pi_{\mu f} = -4h_{\mu f}\sigma_{\mu f},$$

$$H_{\mu f} = \delta_{\mu\sigma}\sum_{\nu=\sigma,\eta} a_\nu h_{\nu f}, \quad a_\mu = \delta_{\mu\sigma} - \delta_{\mu\eta}. \tag{3b}$$

Here $\pi_{\mu f}$ is a QG analog of the reaction Onsager field, introduced in SG theory in Ref. 29; $H_{\mu f}$ stands for the intrinsic quadrupolar Zeeman-type field and is due to the kinematic properties of the dynamic quadrupolar variables:[30] $2S_{\mu f}^2 = 1 + a_\mu S_{\sigma f}$. The mean and variance of the isotropic on the average fluctuation field (3), respectively, h_1 and h_2 have been calculated in Ref. 22. The variance of the effective molecular field components (ε_σ and ε_η) are reestimated here and given by the relationships

$$\varepsilon_\mu(c,T)^2 = \epsilon_2^2\left\{1 - \left(\frac{J}{T}\right)^2 \frac{1-q}{q}[2q(p_\mu - q_\mu) - (1+\kappa^2) \times q_\mu(1-q)]\right\} + \frac{J^4\kappa^2}{8}\left(\frac{1-q}{T}\right)^2\delta_{\mu\sigma}, \tag{4}$$

with $\epsilon_2^2 = \langle \epsilon_{\mu f}^2\rangle_C = J^2 q$ and $J(c) = \Gamma\sqrt{zc}$, where $z=12$ is nearest-neighbor number on the hcp lattice. The second term comes from the reaction-field effects and the last term is caused solely by the quadrupolar intrinsic field. Here new orientational order parameter components are introduced: $q_\mu = \langle\langle S_\mu\rangle_T^2\rangle_C, p_\mu = \langle\langle S_\mu^2\rangle_T\rangle_C$, which obey the relations $q_\sigma + q_\eta = q$ and $p_\sigma + p_\eta = 1$. The fluctuation-field parameter $\kappa(c)$ appears in (4) to describe the random-substitution effects through the random-bond correlation coefficient K_B, namely,

$$\kappa(c) = \frac{h_2(c,T)}{h_1(c,T)} = K_B\left(\frac{1-c}{zc}\right)^{1/2}, \quad K_B = \frac{\sqrt{\langle (J_{ff'}^{\mu\mu'} J_{ff'}^{\nu\nu'})^2\rangle_B}}{\langle (J_{ff'}^{\mu\mu'})^2\rangle_B}. \tag{5}$$

One can expect that for the QG phase bounded in the concentration range $c_0 \leqslant c \leqslant c_M (c_M = 0.55$ and $c_0 \sim 0.1$, see *o*-*p*-H_2 diagrams in Refs. 1 and 3) the QG fluctuation fields introduced through the Hamiltonian (3) should be also restricted, i.e., $\kappa(c_M) \leqslant \kappa(c) \leqslant \kappa(c_0)$. Formally, we introduce the lower critical concentration c_0 as a singular point of the amplitude of the effective molecular field $[\varepsilon(c,T) = (\varepsilon_\sigma^2 + \varepsilon_\eta^2)^{1/2}]$ (4) at which the leading terms of the reaction-field effects disappear in high-T and low-T asymptotics of the QG order parameter. We see that this critical condition is $\kappa(c_0) = 1$ which gives $c_0 = (1 + z/K_B^2)^{-1}$. Adopting a Gaussian estimation for the random-bond correlation coefficient[31] (5), $K_B = \sqrt{3}$, and comparing with the findings of dynamic NMR experiments[5,32,33] for the QG lower critical concentration mentioned above, we conclude that low-T consideration requires accounting of the next-nearest-neighbor interactions.

The self-consistent equations for the orientational order parameter $q(c,T) = \langle \sigma_f^2 + \eta_f^2\rangle_C$ follow from (2) and should be completed by the equation $\sigma(c,T) = \langle \sigma_f\rangle_C$, namely,

$$q(c,T) = 1 - 3\left\langle \frac{1 + 2\exp(ax)\cosh(by)}{[\exp(ax) + 2\cosh(by)]^2}\right\rangle_{x,y},$$

$$a(c,T) = \tfrac{3}{2}\,\varepsilon_\sigma(c,T)/T, \tag{6a}$$

$$\sigma(c,T) = 1 - 3\left\langle \frac{\cosh(by)}{\exp(ax) + 2\cosh(by)}\right\rangle_{x,y},$$

$$b(c,T) = (\sqrt{3}/2)\,\varepsilon_\eta(c,T)/T, \tag{6b}$$

where $\langle\cdots\rangle_{x,y}$ denotes nonsymmetrical Gaussian integration $(x_1 = \varepsilon_1/\varepsilon_\sigma)$ over the local fields $(\varepsilon_{\sigma f} = x\varepsilon_\sigma$ and $\varepsilon_{\eta f} = y\varepsilon_\eta)$. To give low-T consideration in the explicit form[34] we introduce the following asymptotic representations:

$$q(c,T) \sim 1 - 3\langle \varphi_q(x,y)\rangle_{x,y}, \quad T \to 0,$$

with

$$\varphi_q(x,y) = \exp(ax - by)/[1 + \exp(ax - by)]^2, \tag{7a}$$

$$\sigma(c,T) \sim \tfrac{1}{2} + \tfrac{3}{2}\langle \varphi_\sigma(x,y)\rangle_{x,y}, \quad T \to 0,$$

with $\varphi_\sigma(x,y) = \exp(ax - by)/[1 + \exp(ax - by)]$ (7b)

in the range of variables $|x| < \infty$ and $y < \infty$. As a further simplification, we define the following $T \to 0$ asymptotic equivalent functions: $\varphi_q(x,y) \sim \delta(ax - by)$ and $\varphi_\sigma(x,y) \sim \Theta(ax - by)$, with usual notations for the Dirac δ function and the Heaviside step function. After performing integration the order-parameter-equation system can be presented in the explicit form

$$q(c,T) = 1 - \sqrt{3/2\pi}\,T/\varepsilon_{\mathrm{LT}}(c,T) + O(T/J)^3,$$

$$\varepsilon_{\mathrm{LT}} = \tfrac{1}{2}\sqrt{3\varepsilon_\sigma^2 + \varepsilon_\eta^2}, \tag{8a}$$

$$\sigma(c,T) = \frac{1}{4}\left(1 - \frac{\varepsilon_\eta(c,T)}{\varepsilon_\sigma(c,T)}\right) + \frac{3}{4\sqrt{2\pi}}\frac{\varepsilon_1(c,T)}{\varepsilon_\sigma(c,T)} + O\left(\frac{T}{J}\right)^2,$$

$$\varepsilon_1(c,T) = -4\sigma(c,T)h_1(c,T). \tag{8b}$$

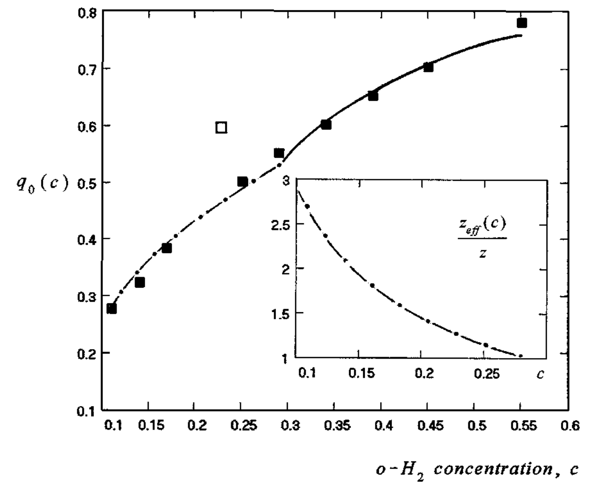

FIG. 1. Quadrupolar glass orientational order parameter against ortho-concentration in ortho-para-hydrogen mixtures. Closed and open points correspond to the extrapolated to zero-temperature NMR data of, respectively, Refs. 14 and 15. Solid and dashed-dotted lines show the solutions of Eq. (10), respectively, in the nearest-neighbor ($z=12$) and effective mean-distance-neighbor ($z_{\rm eff}$) interaction approximations [$q_M=0.72$ and $\kappa(c_M)=0$ are adopted]. Inset: reduced number of the effective mean-distance neighbors $z_{\rm eff}(c)/12=(\bar{R}/R_0)^3$ with $\bar{R}(c)=(3R_0/4\pi\sqrt{2}c)^{1/3}$ and R_0 are, respectively, the mean and nearest-neighbor distances on the hcp lattice.

To illuminate the low-T behavior of the effective MF characteristic energy $\varepsilon_{\rm LT}=\epsilon_2\sqrt{g_{\rm LT}}$ we introduce the reaction-field factor $g_{\rm LT}$ by means of relations

$$g_{\rm LT}(c,T)=1-\frac{J^2(c)}{2}\{[1-q(c,T)]/T\}^2\lambda_{\rm LT}(c,T);$$

$$\lambda_{\rm LT}(c,T)=1-\kappa^2(c)-3\kappa^2(c)/16q(c,T)\ , \tag{9a}$$

which in turn can be formally interpolated to low temperatures:

$$g_{\rm LT}(c,T)=\frac{1}{1+[J^2(c)/2][1-q_0(c)/T]^2\lambda_{\rm LT}(c,0)},$$

$$q_0=q(c,0). \tag{9b}$$

For the MF characteristic energy $\varepsilon_{\rm LT}$ one has the following asymptotics:

$$\varepsilon_{\rm LT}(c,T)\sim T\sqrt{2q_0(c_M)}/[1-q_0(c)],\quad T\rightarrow 0. \tag{9c}$$

Unusual for SG's reaction-field behavior,[35] this leads to the low-T effect of the disappearing of the molecular field (9c). The latter has been derived from NMR data[14] by Li *et al.* (see Fig. 9 of Ref. 16 and recently discussed in Ref. 25. To examine a correspondence with experiment in concentrations and in a way to verify the microscopic description of spatial correlations we have used (9c) to find a nontrivial zero-temperature asymptotic solution for the QG orientational order parameter[36] (8a), namely,

$$q_0^2/q_M-[1-\kappa^2(c)]q_0+\tfrac{3}{16}\kappa^2(c)=0,\quad q_M=q(c_M,0) \tag{10}$$

with $q_M=3/4\pi$ [the solution $\sigma(c,T)\sim T^2$ (8b) is taken into account]. Analysis of (10) and its comparison with experiment is given in Fig. 1.

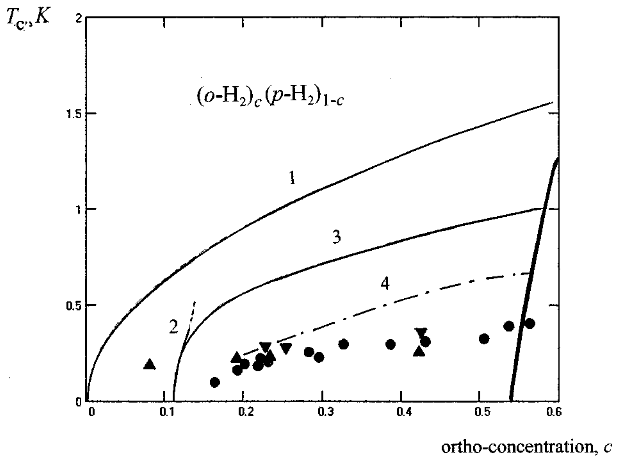

FIG. 2. Phase diagram for ortho-para-hydrogen mixtures. Lines: mean-field estimations for zero-field freezing temperature within the quadrupolar glass vector model: (1) T_F (12a), in the absence or the reaction and instrinsic fields; (2) $\bar{T}_{0F}$ (12a) and (3) T_{0F} (12b) in the presence of the reaction field, respectively; (4) Almeida-Thouless instability line within the quadrupolar glass infinite-range axial model in the presence of the quadrupolar intrinsic field (Ref. 37). Points: the dynamic NMR anomalies observed by Sullivan *et al.* (●) (Ref. 5), Ishimoto *et al.* (▲) (Ref. 32), and Husa and Daunt (▼) (Ref. 33). (See also comments in Ref. 3.)

The high-T asymptotics for the orientational order parameter follows immediately from (6):

$$q(c,T)=\frac{h_{Cr}^2+h_2^2(c,T)}{2T^2}+\frac{\varepsilon_{HT}^2(c,T)}{2T^2}-O\left(\frac{h_{Cr}^2h_2^2(c,T)}{T^4}\right), \tag{11a}$$

$$\varepsilon_{HT}(c,T)=J(c)\sqrt{q}(c,T)\left[1-\frac{J^2(c)}{2}\left(\frac{1-q(c,T)}{T}\right)^2\times[1-\kappa^2(c)]\right]^{1/2}. \tag{11b}$$

In contrast to the low-T case small-amplitude crystalline-field effects (1) should be taken into account.[22] Formally, the first term in (11) is due to "external" random fields. In the spirit of mean-field theory of SG's in an external field one can estimate a zero-field freezing temperature T_{0F}. Omitting formally $h_{\rm Cr}$ and h_2 in (11a) we have the following equation for the freezing temperature T_F now improved by the reaction-field effects:

$$\frac{\bar{T}_{0F}^4}{T_F^2(c)}-\bar{T}_{0F}^2+[1-\kappa^2(c)]\bar{T}_F^2(c)=0,\quad T_F(c)=\Gamma\sqrt{zc}/2. \tag{12a}$$

This square equation has a nontrivial solution, which for instructiveness can be approximated as $\bar{T}_{0F}(c)\sim\Gamma\sqrt{c-c_0}$. It holds in the narrow concentration range, $c_0\leqslant c\leqslant 5c_0/4$, where the value $[1-\kappa^2(c)]$ is small. Considering formally the latter as a small parameter the iteration corrections to the last term in (12a) have been found in all orders of its magnitude to extend the applicability of the solution of Eq. (12a) to the whole QG concentration range, namely,

$$T_{0F}^4/T_F^2(c)-T_{0F}^2+\left(1+\frac{1}{1-\kappa^2(c)}\,\frac{\bar{T}_{0F}^2(c)}{T_F^2(c)}\right)^{-1}\bar{T}_{0F}^2(c)=0, \tag{12b}$$

where $\bar{T}_{0F}^2(c)$ stands for one of the solutions of the first-order-approximation Eq. (12a). Both solutions are illustrated in Fig. 2. In the next step one should include the ‘‘external’’ fields mentioned above to reveal the para-rotational phase instability[37] in the spirit of that obtained for SG's in a constant external field.[38]

The microscopic analysis of the competing ordering fields given above permits one to conclude that the glasses under discussion are characteristically different rather in the local reaction-polarization effects that in the kinematic properties of their dynamic variables. We summarize the main results of the microscopical approach to the QG problem as follows. (1) Orientational ordering in QG's has a collective character and is not caused by the intrinsic quadrupolar field as earlier suggested in Ref. 3. In contrast to SG's where the reaction-field effects can be reduced to corrections (see, e.g., Ref. 39 and discussion in Ref. 25), the QG orientational freezing is due to the correlated reaction and molecular fields. (2) Nonzero variance of the fluctuation field specific of QG systems gives rise to incomplete low-temperature orientational ordering (Fig. 1). (3) The QG phase formation requires a certain threshold concentration c_0 (Fig. 2), above which the variance of the fluctuation field is relatively small. (4) The quadrupolar intrinsic field dominates at high temperatures and can be self-consistently treated as an effective external field.

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[35] The QG reaction-field results given in (4), (9a), and (11b) can be compared with the *random-bond* SG case of the $S=\frac{1}{2}$ Ising model, namely, $\varepsilon_{\mathrm{SG}}(T)=\epsilon_2(T)(1+J^2\{[1-q(T)]/T\}^2)^{1/2}$, with $\epsilon_2(T)=J\sqrt{q(T)}$. One can speculate on its qualitative correspondence with the QG case at $c<c_0$.

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