Equilibrium properties of a quadrupolar glass

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(Received 1 March 1994)

We introduce a semimicroscopic discrete-state model appropriate to the orientational glass phase in mixed alkali halide cyanides with ⟨111⟩ equilibrium orientations of the CN^- ions, such as $K(CN)_xBr_{1-x}$. The order-parameter fields are defined as symmetry adapted combinations of the occupation number operators along the cubic body diagonals, which transform according to the T_{2g} representation of the cubic group. These interact via an infinite-range random interaction in the presence of quenched local random strains. We then use the replica formalism to derive a replica-symmetric solution for the components of the orientational-glass order parameter, the linear susceptibilities, and the elastic compliances. The high-temperature orientational-glass phase is characterized by an isotropic order-parameter matrix with only the diagonal elements q_μ being nonzero. At high temperatures, the behavior of the order parameter $q = \sum_\mu q_\mu/3$ is similar to that of an Ising spin glass, however, at intermediate and low temperatures the two models differ significantly. We also derive the instability line $T_f(\Delta)$ separating the replica-symmetric isotropic phase from the low-temperature anisotropic orientational glass phase, which is characterized by broken replica symmetry. In contrast to the random-bond–random-field model of an Ising spin glass, the instability temperature increases with random-field variance, implying that in quadrupolar glasses replica-symmetry breaking may be relevant already at relatively high temperatures. Finally, an expression for the distribution of local strains related to the NMR line shape is derived. It is also shown that the quadrupolar glass order parameter can be determined by NMR.

I. INTRODUCTION

Orientational-glass ordering which occurs in mixed alkali halide cyanides such as $K(CN)_xBr_{1-x}$ is characterized by frozen-in random orientations of the quadrupolar axis of CN^- ions. There is a large body of experimental data regarding the equilibrium and dynamic properties of these and similar crystals,[1] in which the nonspherical CN^- ion is randomly substituted by a spherical ion, e.g., Br^-.

In quadrupolar glasses of the above type, the interaction between the quadrupoles is mediated by the elastic deformations of the fcc center-of-mass lattice. The sign of the pair interaction energy depends on both the mutual orientation of the quadrupole tensors and their orientation with respect to the bond vector. In a disordered system, the signs are expected to alternate randomly, implying that the interactions are frustrated, and as a result the quadrupolar axes freeze in random discrete orientations. Thus an orientational-glass phase appears for concentrations x below a threshold value x_c, which for $K(CN)_xBr_{1-x}$ is found to be close to 0.7.[2] For $0.7 < x < 0.9$ a ferroelastic first-order phase transition from the cubic to a rhombohedral phase occurs. For $0.9 < x < 1$ another first-order cubic-to-orthorhombic phase transition characterized by a ferroelastic long-range order occurs, similar to the one in pure KCN.[2] In $K(CN)_xBr_{1-x}$ the phase diagram is strongly asymmetric with respect to x; i.e., the glassy phase is found down to very low CN^- concentrations with progressively smaller values of the transition temperature. Ultrasonic measurements[3] in the glassy phase suggest that the lattice remains effectively cubic down to the lowest temperatures. The transition into the glass phase is associated with an anomaly in the shear compliance. The freezing temperature T_f can be estimated from the experiments of Hessinger and Knorr,[4] who observed the splitting between the field-cooled and zero-field-cooled linear elastic susceptibilities in $K(CN)_xBr_{1-x}$. It was further found by NMR on a closely related system $Na_xK_{1-x}CN$ (Ref. 5) that above the freezing temperature the NMR lines are broadened, indicating that the orientational-glass order parameter remains finite at all temperatures, similar to the dipolar glasses[6] where the presence of local random fields was found to be responsible for the appearence of noncooperative order above the nominal transition temperature. Analogous conclusions were also reached by neutron scattering experiments[7] on $K(CN)_xBr_{1-x}$. Thus random local strains appear to be an essential feature which should be included into any realistic model of quadrupolar glasses.[8]

By measuring the nonlinear elastic susceptibility χ_{NL} of $K(CN)_xBr_{1-x}$, Hessinger and Knorr[9] found a scaling behavior $\chi_{NL} \sim (T - T_{NL})^{-\gamma}$, where T_{NL} roughly coincides with the freezing temperature T_f. This gives a strong indication that random interactions do exist in

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these systems along with random strain fields, since the latter alone could not produce a diverging nonlinear response. Below T_f a remanent strain was observed,[4] which disappears on approaching T_f, in full analogy with the remanent magnetization in magnetic spin glasses.[10] Moreover, the time dependence of the remanent strain shows[4] a nonexponetial decay, which again is a characteristic feature of spin glasses and related systems.

Theoretical concepts relevant to the subject of orientational-glass ordering and the results of numerical simulations on these systems are summarized in a recent review by Binder and Reger.[11] In general, two classes of models have been studied: (i) models characterized by continuous orientation of the quadrupolar axis,[12] and (ii) discrete-state models.[13] The complexity of a theoretical description of the orientational-glass phase lies in the fact that in general the fluctuation field is a m-dimensional tensor of rank 2. For instance, in the case of continuous symmetry models with m=3, the orientational-glass order parameter is a 6×6 matrix. In mixed alkali halide cyanides, however, the CN^- quadrupoles cannot rotate freely due to the strong hindering potential generated by the neighboring ions. Thus the crystal-field anisotropy will invariably reduce the number of possible orientations of the quadrupolar axis to a discrete set of states, which correspond to the minima of the anisotropy potential on a mezoscopic scale.[13] In a cubic crystal these can lie along the cubic axes ⟨100⟩, body diagonals ⟨111⟩, or face diagonals ⟨110⟩. As shown by Vollmayr *et al.*[13] (to be referred to as VKZ), the discrete-state equilibrium orientations of the quadrupolar axis along the ⟨100⟩, ⟨111⟩, and ⟨110⟩ directions can be described by the s-state Potts model, with s = 3, 4, and 6, respectively.

In the present work we focus on a discrete-state model appropriate to the case of ⟨111⟩ equilibrium orientations of the quadrupolar axis. We investigate the orientational-glass phase for a range of concentrations x just below the threshold value $x_c \approx 0.7$, i.e., in the vicinity of the ferroelastic rhombohedral phase, where the sample-averaged shear strains disappear; however, they are expected to be locally nonzero giving rise to local random strain fields. We start from the VKZ (Ref. 13) representation of discrete occupation numbers N_{ip}, where $p = 1, 2, 3, 4$ labels the four ⟨111⟩ states. In contrast to earlier models, however, we explicitly include at the outset both an isotropic random-bond interaction of the VKZ type and a set of local random strain fields. In general, local random strains can be represented by their irreducible symmetry components, which couple linearly to the order parameter field. Thus a set of symmetry-adapted linear combinations of N_{ip} appears as a natural choice for the appropriate order parameter fields, which—like the random strains themselves—transform according to the T_{2g} representation of the cubic group. An extension of the present approach to the cases of ⟨100⟩ and ⟨110⟩ equilibrium orientations appears straightforward, but is rather tedious in view of the algebra of the appropriate order-parameter fields, and will be discussed in a future publication.

The organization of the paper is as follows: In Sec. II we introduce the model and by using the replica theory derive an expression for the averaged free energy. In Sec. III, the replica-symmetric solution is obtained and the temperature dependences of the orientational-glass order parameter, the linear susceptibilities, and the shear compliances are calculated numerically. We also derive an analytic expression for the local shear strain distribution. In Sec. IV we investigate the stability of the isotropic symmetric solution against replica-symmetry-breaking fluctuations. Finally, Sec. V is devoted to a discussion and a short summary of the results.

II. DISCRETE-STATE MODEL AND MEAN-FIELD THEORY

A theoretical model capable of describing the phase transition into the orientational-glass phase in $K(CN)_xBr_{1-x}$ crystals can be set up by first introducing the orientational degrees of freedom—i.e., the orientations of the uniaxial-quadrupole axis—which are linearly coupled to elastic strains. On a semimicroscopic scale the lattice remains cubic in the orientational-glass phase. For simplicity, we will assume that each lattice site is occupied by a discrete-state quadrupole, and that these are interacting via randomly frustrated long-range interactions. For concentrations x in the vicinity of the ferroelastic rhombohedral phase, the predominant quadrupolar axis orientations are along the cubic body diagonals ⟨111⟩. In this case the orientational degrees of freedom can be adequately represented by the occupation numbers N_{ip}, where p=1, 2, 3, 4 labels the cubic body diagonals at lattice site i. By definition, N_{ip}=1 or 0 and thus $(N_{ip})^2 = N_{ip}$. Following VKZ we assume that the interaction J_{ij} is isotropic in the discrete-state representation; i.e., we write the quadrupolar Hamiltonian including a random-field term in the form

$$\mathcal{H}_Q = -\frac{1}{2}\sum_{ij}\sum_{p=1}^{4} J_{ij}N_{ip}N_{jp} - \sum_{i}\sum_{p=1}^{4}\sum_{\mu} h_{i\mu}A_{\mu p}N_{ip}. \tag{1}$$

Here J_{ij} are random interactions and $h_{i\mu}$ are random fields due to local random strains, which are generated by the disorder,[8] with $A_{\mu p}$ representing a set of dimensionless coupling coefficients. In general, the components $h_{i\mu}$ will transform according to the irreducible representations Γ of the cubic O_h group. In the present problem, however, the occupation numbers N_{ip} restrict the space of $\mu\{\Gamma\}$ to A_{1g} and T_{2g} representations only. Thus we may introduce a set of symmetry adapted variables $Z_{i\mu}$ at each lattice site, namely,

$$Z_{i\mu} = \sum_{p} A_{\mu p}N_{ip}\,, \tag{2}$$

where the matrix $A_{\mu p}$ is given by the following explicit relations:

$$Z_1 = N_1 + N_2 - N_3 - N_4\,, \tag{3}$$
$$Z_2 = N_2 + N_3 - N_1 - N_4\,, \tag{4}$$
$$Z_3 = N_3 + N_1 - N_2 - N_4\,, \tag{5}$$
$$Z_4 = N_1 + N_2 + N_3 + N_4\,. \tag{6}$$

Here we have dropped the site indices i. Clearly, the A_{1g} component is $Z_4 = 1$ in view of the closure relation $\sum_p N_{ip} = 1$. Thus Z_1, Z_2, Z_3 are the only three nontrivial components of the order parameter, which transform according to the T_{2g} representation. The algebra of $Z_\mu, \mu = 1,2,3$, is from Eqs. (3)–(5) given by

$$Z_\mu Z_\nu = \delta_{\mu\nu} - Z_\lambda(1-\delta_{\mu\lambda})(1-\delta_{\nu\lambda})(1-\delta_{\mu\nu}) . \quad (7)$$

It may also be noted that $\sum_{\mu=1}^{3} Z_\mu = N_1 + N_2 + N_3 - 3N_4 \neq 1$.

In terms of Z_μ variables the orientational part of the Hamiltonian can be rewritten as

$$\mathcal{H}_\mathcal{Q} = -\frac{1}{2}\sum_{ij}\sum_{\mu=1}^{3} J_{ij} Z_{i\mu} Z_{j\mu} - \sum_i \sum_{\mu=1}^{3} h_{i\mu} Z_{i\mu} . \quad (8)$$

The total Hamiltonian

$$\mathcal{H}_\mathcal{T} = \mathcal{H}_\mathcal{Q} + \mathcal{H}_\mathcal{I} + \mathcal{H}_\mathcal{E} \quad (9)$$

includes in addition to the quadrupolar Hamiltonian (8) the elastic energy $\mathcal{H}_\mathcal{E}$ due to propagating acoustic waves, and the interaction between the acoustic waves and quadrupoles, $\mathcal{H}_\mathcal{I}$. These are given by the following two expressions:

$$\mathcal{H}_\mathcal{E} = v_0 \sum_i \left[\frac{1}{2}(C_{11}+2C_{12})e_{i1}^2 + (C_{11}-C_{12})(e_{i2}^2 + e_{i3}^2) + \frac{1}{4}C_{44}(e_{i4}^2 + e_{i5}^2 + e_{i6}^2) \right] , \quad (10)$$

$$\mathcal{H}_\mathcal{I} = \gamma \sum_i \sum_{\mu=1}^{3} Z_{i\mu} e_{i,\mu+3} . \quad (11)$$

Here v_0 is the unit cell volume and $e_{i\mu}$ are the corresponding symmetry components of the strain tensor ϵ at site i. In the present case, only the shear components $e_4 = \epsilon_{xy}$, $e_5 = \epsilon_{xz}$, and $e_6 = \epsilon_{yz}$ couple to Z_1, Z_2, and Z_3, respectively, all with the same coupling constant γ.

The third-order and higher anharmonic terms not included in $\mathcal{H}_\mathcal{E}$ are not expected to play an important role for the orientational-glass transition, which according to the experimental results remains continuous, in contrast to the ferroelastic transition which is first order.[2] Obviously, in the present model only the elastic constant C_{44} will be directly affected by the orientational-glass ordering. In general, however, third-order terms in $\mathcal{H}_\mathcal{E}$ do exist and will lead to a weaker softening of the elastic constant $C_{11} - C_{12}$.[14]

A. Mean-field theory of the orientational glass

In order to discuss the equilibrium properties of the orientational glass phase we first apply the replica formalism to the subsystem described by the Hamiltonian (8), ignoring for the moment the interactions with the elastic degrees of freedom. The effects of orientational freezing on the acoustic properties will be discussed in Sec. III.

Within the standard replica approach, one writes down the averaged partition function of the n-replicated Hamiltonian and subsequently takes the $n \to 0$ limit in order to find the free energy as

$$\mathcal{F} = -\frac{1}{\beta}\lim_{n\to 0}\frac{[\mathcal{Z}^n]_{\rm av} - 1}{n}, \quad (12)$$

where the averaged partition function is

$$[\mathcal{Z}^n]_{\rm av} = \left[{\rm Tr}_n \exp\left(\frac{1}{2}\beta \sum_\alpha \sum_{ij,\mu} J_{ij} Z_{i\mu}^\alpha Z_{j\mu}^\alpha + \beta \sum_\alpha \sum_{i\mu} h_{i\mu} Z_{i\mu}^\alpha \right) \right]_{\rm av} , \quad (13)$$

and $[\cdots]_{\rm av}$ denotes a joint random average over the probability distributions of random bonds J_{ij} and random fields $h_{i\mu}$. As usual, we assume that these distributions are Gaussian and uncorrelated. Their first and second moments are given by $[J_{ij}]_{\rm av} = J_0/N$ and $[J_{ij}^2]_{\rm av} = J/\sqrt{N}$ for random bonds, and by $[h_{i\mu}]_{\rm av} = 0$ and $[h_{i\mu}h_{j\nu}]_{\rm av} = \Delta\delta_{ij}\delta_{\mu\nu}$ for random fields, respectively. The trace in Eq. (13) is over all possible values of Z_μ^α. After performing the Gaussian averages in Eq. (13) we have apart from a constant:

$$[\mathcal{Z}^n]_{\rm av} = {\rm Tr}_n \exp\left[\frac{\beta J_0}{2N}\sum_{\mu\alpha}\left(\sum_i Z_{i\mu}^\alpha\right)^2 + \frac{\beta^2 J^2}{4N}\sum_{\mu\nu}\sum_{\alpha\beta}\left(\sum_i Z_{i\mu}^\alpha Z_{i\nu}^\beta\right)^2 + \frac{\beta^2\Delta}{2}\sum_{i\mu}\sum_{\alpha\beta} Z_{i\mu}^\alpha Z_{i\mu}^\beta \right] . \quad (14)$$

The quadratic forms in the above expression can be decomposed in order to achieve an effective single-site Hamiltonian by introducing two sets of field variables,

$$\hat{q}_{\mu\nu}^{\alpha\beta}(Z) = \frac{1}{N}\sum_i Z_{i\mu}^\alpha Z_{i\nu}^\beta \ , \ \alpha \neq \beta \ , \quad \hat{P}_\mu^\alpha(Z) = \frac{1}{N}\sum_i Z_{i\mu}^\alpha . \quad (15)$$

Then the trace in Eq. (14) can be evaluated by constraining the above expressions to constant values $\hat{q}_{\mu\nu}^{\alpha\beta}$ and $\hat{P}_\mu^\alpha$, respectively, and integrating over all possible paths of $\hat{q}_{\mu\nu}^{\alpha\beta}(Z)$ and $\hat{P}_\mu^\alpha(Z)$. The constraints are imposed by introducing

two sets of Lagrange multipliers $r_{\mu\nu}^{\alpha\beta}$ and λ_μ^α. Using the appropriate measures $\mathcal{D}[\lambda_\mu]$, $\mathcal{D}[r_{\mu\nu}]$, $\mathcal{D}[\hat{P}_\mu]$, and $\mathcal{D}[\hat{q}_{\mu\nu}]$ we can write

$$[\mathcal{Z}^n]_{\rm av} = \int \mathcal{D}[\hat{P}_\mu] \int \mathcal{D}[\lambda_\mu] \int \mathcal{D}[\hat{q}_{\mu\nu}] \int \mathcal{D}[r_{\mu\nu}] \exp\left[-N \sum_{\mu\alpha} \left(\lambda_\mu^\alpha \hat{P}_\mu^\alpha - \frac{1}{2}\beta J_0' (\hat{P}_\mu^\alpha)^2\right)\right]$$
$$\times \exp\left[-N \sum_{\mu\nu} \sum_{\alpha\neq\beta} \left(r_{\mu\nu}^{\alpha\beta} \hat{q}_{\mu\nu}^{\alpha\beta} - \frac{1}{4}\beta^2 J^2 (\hat{q}_{\mu\nu}^{\alpha\beta})^2\right) + \frac{3}{4}\beta^2 J^2 nN\right.$$
$$\left. + \ln {\rm Tr}_n \exp\left(\sum_{\mu\alpha} \sum_i \lambda_\mu^\alpha Z_{i\mu}^\alpha + \sum_{\mu\nu} \sum_{\alpha\neq\beta} \sum_i r_{\mu\nu}^{\alpha\beta} Z_{i\mu}^\alpha Z_{i\nu}^\beta + \frac{\beta^2\Delta}{2} \sum_{\alpha\beta} \sum_{i\mu} Z_{i\mu}^\alpha Z_{i\mu}^\beta\right)\right] . \quad (16)$$

In the limit $N \to \infty$, the integrations over $\hat{q}_{\mu\nu}$, $r_{\mu\nu}$, $\hat{P}_\mu$, and λ_μ can be evaluated using saddle-point methods, which is equivalent to replacing $\hat{q}_{\mu\nu}$, $r_{\mu\nu}$, $\hat{P}_\mu$, and λ_μ by their stationary values. These are

$$q_{\mu\nu}^{\alpha\beta} = \frac{1}{N} \sum_i \langle Z_{i\mu}^\alpha Z_{i\nu}^\beta \rangle , \quad (17)$$

$$P_\mu^\alpha = \frac{1}{N} \sum_i \langle Z_{i\mu}^\alpha \rangle , \quad (18)$$

representing the order parameters in the problem, and

$$r_{\mu\nu}^{\alpha\beta} = \frac{1}{2}\beta^2 J^2 q_{\mu\nu}^{\alpha\beta} , \quad (19)$$

$$\lambda_\mu^\alpha = \beta J_0' P_\mu^\alpha , \quad (20)$$

where

$$J_0' = J_0 + \beta J^2 . \quad (21)$$

By inserting the saddle-point values (19) and (20) back into Eq. (16) and returning to Eq. (12), we finally obtain the formal result for the free energy per particle $f \equiv \beta\mathcal{F}/N$:

$$f = -\frac{3}{4}\beta^2 J^2 - \lim_{n\to 0} n^{-1} \max_{\{P,q\}} \left\{ -\frac{1}{2}\beta J_0' \sum_{\mu\alpha} (P_\mu^\alpha)^2 - \frac{1}{4}\beta^2 J^2 \sum_{\mu\nu} \sum_{\alpha\neq\beta} \left(q_{\mu\nu}^{\alpha\beta}\right)^2 \right.$$
$$\left. + \ln {\rm Tr}_n \exp\left(\beta J_0' \sum_{\mu\alpha} Z_\mu^\alpha P_\mu^\alpha + \frac{1}{2}\beta^2 J^2 \sum_{\mu\nu} \sum_{\alpha\neq\beta} \left(q_{\mu\nu}^{\alpha\beta} + \tilde{\Delta}\delta_{\mu\nu}\right) Z_\mu^\alpha Z_\nu^\beta\right)\right\} . \quad (22)$$

Here we have introduced the notation $\tilde{\Delta} \equiv \Delta/J^2$.

Using the algebra of Z_μ [cf. Eq. (7)], the terms in Eq. (16) proportional to $\beta^2 J^2$ with $\alpha = \beta$ have been combined with the quadratic term proportional to βJ_0, thus renormalizing the interaction parameter J_0 according to Eq. (21). In the following we will consider the case $|J_0'| < J$, implying that long-range order is absent. As discussed later, the above condition is satisfied in the range of temperatures and concentrations which is of interest experimentally.

III. ISOTROPIC REPLICA-SYMMETRIC ORIENTATIONAL-GLASS PHASE

In this section we search for the replica-symmetric solution for the orientational-glass order parameter by assuming that for all pairs $\alpha \neq \beta$,

$$q_{\mu\nu}^{\alpha\beta} = q_\mu \delta_{\mu\nu} . \quad (23)$$

The off-diagonal components $q_{\mu\nu}^{\alpha\beta}$ with $\mu \neq \nu$ must be zero, since they are not invariant with respect to a set of gauge transformations in which the indices of N_p in Eqs. (3)–(5) are relabeled in all possible manners. Inserting the ansatz (23) into Eq. (22) and using standard Gaussian transformations, we find that apart from a constant, the free energy per site is given by

$$f = -\frac{1}{4}\beta^2 J^2 \sum_\mu (q_\mu - 1)^2 + \frac{1}{2}\beta J_0' \sum_\mu P_\mu^2 - \int \prod_\mu \left[\frac{dx_\mu}{\sqrt{2\pi}} e^{-\frac{1}{2}\sum_\mu x_\mu^2}\right] \ln {\rm Tr} \exp\left(\beta \sum_\mu H_\mu Z_\mu\right) , \quad (24)$$

where the trace is to be taken over the Z_μ variables, and the effective field H_μ acting on Z_μ is given by

$$H_\mu = J\sqrt{q_\mu + \tilde{\Delta}}\, x_\mu + J_0' P_\mu + E_\mu^{\rm ext} . \quad (25)$$

Here x_μ is the component of the Gaussian static field induced by a nonzero orientational-glass order parameter q_μ and random-field variance $\tilde{\Delta}$, and E_μ^{ext} is the conjugate field to Z_μ formally introduced to generate the static response functions.

Calculating the trace in Eq. (24) we finally obtain the following expression for the free energy in the replica symmetric case:

$$f = -\frac{1}{4}\beta^2 J^2 \sum_\mu (q_\mu - 1)^2 + \frac{1}{2}\beta J_0' \sum_\mu P_\mu^2 - \left[\ln\left(4\cosh(\beta H_1)\cosh(\beta H_2)\cosh(\beta H_3)\right) + \ln\left(1 - \tanh(\beta H_1)\tanh(\beta H_2)\tanh(\beta H_3)\right)\right]_{123}, \tag{26}$$

where $[\cdots]_{123}$ denotes a triple Gaussian average over the fields x_μ, $\mu = 1, 2, 3$, and H_μ is given by Eq. (25).

A. Orientational-glass order parameter and linear susceptibilities

We now obtain equations for the "polarizations" P_μ and components of the orientational-glass order parameter q_μ in the replica-symmetric phase by imposing the corresponding extremal conditions on the free energy functional (26). This leads to the following expressions:

$$P_\mu = \left[p_\mu\right]_{123} \tag{27}$$

and

$$q_\mu = \left[p_\mu^2\right]_{123}, \tag{28}$$

where p_μ is the local "polarization"—corresponding physically to a shear strain—which is given by

$$p_\mu = \frac{\tanh(\beta H_\mu) - \prod_{\nu\neq\mu}\tanh(\beta H_\nu)}{1 - \prod_\nu \tanh(\beta H_\nu)}. \tag{29}$$

As before, $[\cdots]_{123}$ stands for the average over three Gaussian fields x_1, x_2, x_3. The corresponding linear susceptibility χ_μ is determined as

$$\chi_\mu = \left(\frac{\partial P_\mu}{\partial E_\mu^{\text{ext}}}\right)_{E_\mu^{\text{ext}}=0}. \tag{30}$$

From the above expressions it follows that in the case $J_0' = 0$ one has

$$\chi_\mu = \beta\left[\langle Z_\mu^2\rangle - \langle Z_\mu\rangle^2\right]_{123} = \beta(1 - q_\mu). \tag{31}$$

We may also define a scalar order parameter q (Ref. 11) as the sum over all components $q_{\mu\nu}$, namely,

$$q = \frac{1}{3}\sum_\mu q_\mu. \tag{32}$$

From the expressions (28) and (29) we see that in the replica-symmetric phase and in the absence of external fields all three components of the order-parameter matrix are equal, thus leading to an isotropic glass phase with $q_1 = q_2 = q_3 = q$. In Fig. 1 we plot the order parameter q as a function of reduced temperature T/J for various values of the reduced random-field variance $\tilde{\Delta}$. Here we have assumed that the effective first moment of the distribution of random interactions J_0' vanishes. (See Sec. V for a more detailed discussion of the conditions under which long-range order is suppressed.) In Fig. 2 the linear susceptibility $\chi_\mu = \chi = \beta(1 - q)$ is plotted against T/J for the same values of $\tilde{\Delta}$. For comparison, we also plot q_I and χ_I, i.e., the spin-glass order parameter and linear susceptibility, respectively, obtained for the random-bond–random-field (RB-RF) Ising model[6] with $\tilde{\Delta} = 0.01$ (dotted curves in Figs. 1 and 2). It is easy to see that at high temperatures the orientational-glass order parameter q and the linear susceptibility χ are very well approximated by the corresponding quantities obtained in the case of Ising spins.[6] This justifies use of the Ising RB-RF model in analyzing the experimental data obtained by NMR measurements at high temperatures.[15] At lower temperatures, however, the difference between these two cases is significant.

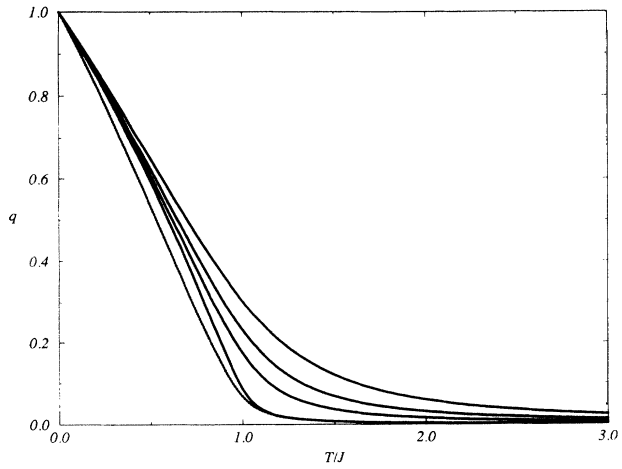

FIG. 1. Orientational-glass order parameter q in the isotropic replica-symmetric phase plotted against reduced temperature T/J for various values of the random-field variance Δ/J^2 (solid lines top to bottom) 0.2, 0.1, 0.05, and 0.01, and for the RB-RF Ising model (dotted line) for $\Delta/J^2 = 0.01$.

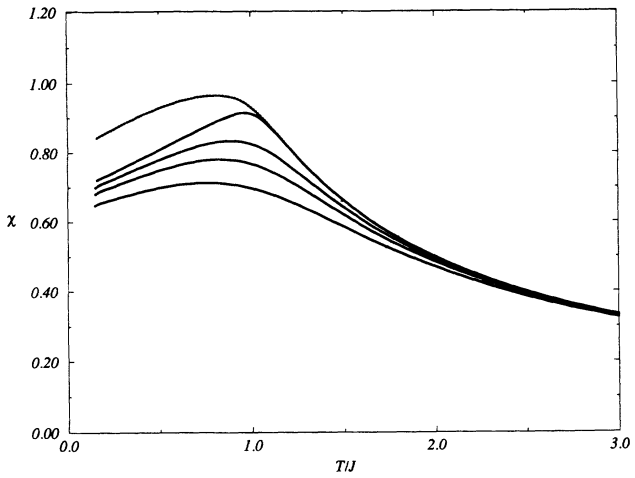

FIG. 2. Linear susceptibility in units of J plotted vs T/J for various values of Δ/J^2 (solid lines top to bottom) 0.01, 0.05, 0.1, and 0.2 . Also plotted is the linear susceptibility for RB-RF Ising model (dotted line) for $\Delta/J^2 = 0.01$.

B. Elastic anomalies

The orientational-glass ordering discussed above affects the elastic properties of the lattice leading to an anomaly in the temperature dependence of the elastic constant C_{44} due to a coupling between the orientational degrees of freedom and shear elastic strains [see Eqs. (9)–(11)]. Within the framework of a mean-field theory for the orientational-glass phase, we may write the effective single-site Hamiltonian involving the orientational degrees of freedom as

$$\mathcal{H}_Q^{\text{eff}} = -\sum_{i\mu} H_\mu Z_{i\mu} , \tag{33}$$

where H_μ is the effective field acting on $Z_{i\mu}$ as given by Eq. (25). Then the free energy associated with the total Hamiltonian which is appropriate for a quantitative evaluation of the elastic anomaly of C_{44} becomes

$$\beta\mathcal{F} = -\ln \mathrm{Tr} \exp\Big(-\beta\mathcal{H}_Q^{\text{eff}} - \beta\mathcal{H}_\mathcal{E} - \beta\gamma \sum_i \sum_{\mu=1}^{3} e_{i\mu+3} Z_{i\mu} - \beta v_0 \sum_i \sum_{\mu=4}^{6} \sigma_\mu^{\text{ext}} e_{i\mu} \Big) . \tag{34}$$

As mentioned earlier, within our model only the shear strains are coupled to the orientational degrees of freedom Z_μ. We may further assume that the coupling constant γ is the same for all three strains e_4,e_5,e_6. In the symmetry representation, the elastic part of the Hamiltonian $\mathcal{H}_\mathcal{E}$, which is given by Eq. (10), is diagonal. Therefore, we may write for the elastic compliance S_μ

$$S_\mu = \frac{\partial\langle e_\mu\rangle}{\partial\sigma_\mu^{\text{ext}}} = \frac{1}{V}\frac{\partial}{\partial\sigma_\mu^{\text{ext}}}\left(\frac{\partial\mathcal{F}}{\partial\sigma_\mu^{\text{ext}}}\right) = \beta V\left(\langle e_\mu^2\rangle - \langle e_\mu\rangle^2\right) . \tag{35}$$

Evaluating the last expression by using Eq. (34), we have

$$S_\mu = \frac{1}{C_\mu}\left(1 + \beta\frac{\gamma^2}{v_0 C_\mu}\left[\langle Z_\mu^2\rangle - \langle Z_\mu\rangle^2\right]_{123}\right) . \tag{36}$$

Thus by virtue of Eq. (31) we finally obtain for the relevant elastic constant $C_{44}(T)$:

$$C_{44}(T) = \frac{C_{44}^0}{1 + \tilde{\gamma}^2\beta(1-q)} , \tag{37}$$

where C_{44}^0 is the value of the shear elastic constant far above the transition temperature, and $\tilde{\gamma}^2 = \gamma^2/v_0 C_{44}^0$ defines the reduced coupling constant, which appears as a phenomenological parameter in our model. In Fig. 3 the reduced elastic constant C_{44}/C_{44}^0 is plotted versus reduced temperature T/J for $\tilde{\Delta} = 0.01$ and various values of $\tilde{\gamma}$. For $T/J \gg 1$, the behavior of the elastic constant is determined by the asymptotic behavior of the order parameter, i.e., $q(T) \sim 1/T^2$. At low temperatures, however, $q(T)$ differs significantly from the T^{-2} law (see Fig. 1) and consequently $C_{44}(T)$ cannot be described by the corresponding asymptotic expression, as already found experimentally.[3]

C. Local-strain distribution and NMR line shape

In the isotropic orientational-glass phase, long-range ferroelastic order is absent—i.e., the order parameters (27) vanish—for a range of parameters such that $|J_0'|/J \leq 1$. However, the local shear strains p_μ given by the expression (29) are nonzero, i.e., $p_\mu \neq 0$, $\mu = 1,2,3$, since they are induced by the local fields x_μ. As Eq. (29) suggests, if two of the local strains p_μ are nonzero, then the third one is automatically generated.

The probability distribution $W(p_1,p_2,p_3)$ of local

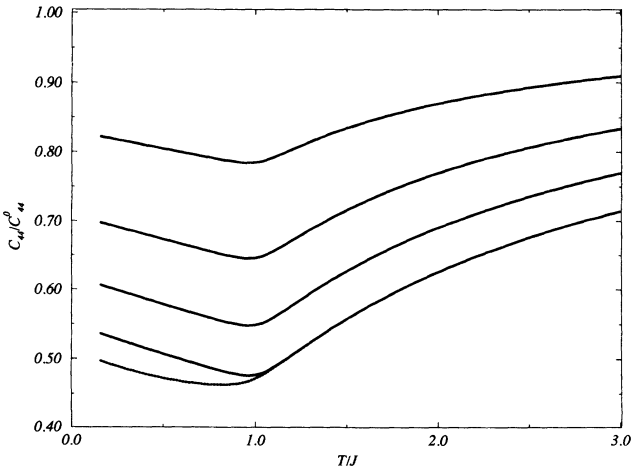

FIG. 3. Shear modulus C_{44}/C_{44}^0 plotted vs T/J for $\tilde{\Delta} = 0.01$ and various values of the reduced coupling parameter $\tilde{\gamma}/J$= 0.3, 0.6, 0.9, and 1.2 (solid lines top to bottom) and for RB-RF Ising model for $\tilde{\Delta} = 0.01$ and $\tilde{\gamma}/J = 1.2$ (dotted line).

strains p_μ is an important quantity which characterizes the orientational-glass phase. In the isotropic replica-symmetric glass phase, $W(p_1,p_2,p_3)$ is implicitly given by a triple Gaussian distribution of static fields x_1,x_2,x_3. We may write

$$W(p_1,p_2,p_3)dp_1dp_2dp_3 = \frac{1}{(2\pi)^{3/2}}\exp\left(-\frac{1}{2}\sum_\mu x_\mu^2\right)dx_1dx_2dx_3 \ . \quad (38)$$

Equation (38) implies that the second moments of $W(p_1,p_2,p_3)$ determine the components of the orientational-glass order parameter q_κ via

$$\int\prod_\mu dp_\mu\, W(p_1,p_2,p_3)p_\kappa^2 \equiv \left[p_\kappa^2\right]_{123} = q_\kappa \ , \quad (39)$$

in full analogy to the Ising spin glass. The relation between the local strain distribution and NMR line shape is, however, not trivial in the present case where more than two different orientational states are involved. Let us assume the following relation between the NMR frequency ν of the ^{14}N nucleus in a CN^- group and the local strains p_μ:

$$\nu = \nu_0 + \nu_1\sum_\mu \alpha_\mu p_\mu \ , \quad (40)$$

where the coefficients α_μ determine the shift of the Larmor frequency due to the electric field gradient tensor, which depends on the orientation of the CN^- ion. The above relation is valid in the fast motion limit where the motion between the discrete orientations of CN^- quadrupoles is fast as compared to the difference in the Larmor frequencies between these sites. Thus the NMR line shape—i.e., the number of lines per unit frequency interval—is given by

$$I_Q(\nu) = \mathrm{Re}\int\prod_\kappa dp_\kappa\, W(p_1,p_2,p_3) \times\delta\left(\nu-\nu_0-\nu_1\sum_\mu\alpha_\mu p_\mu\right) \ . \quad (41)$$

It follows from the above expression and Eq. (39) that the second moment of the NMR line shape (41) is proportional to the orientational-glass order parameter q defined in Eq. (28), namely,

$$M_2 \equiv \int I_Q(\nu)\nu^2 d\nu = q\nu_1^2\sum_\mu\alpha_\mu^2 \ . \quad (42)$$

The coefficients α_μ can be calculated for each particular orientation of the external magnetic field. For instance, when the field is along $\langle 111\rangle$ we have

$$\alpha_1 = -\alpha_2 = -\alpha_3 = 2/3 \ . \quad (43)$$

The dependence of M_2 on the direction of the magnetic field due to the α_μ^2 terms is expected to be relevant at low temperatures, where the calculated NMR line shape differs from the corresponding result for the Ising model,[16] in which case only two different sites of the Ising pseudospin are involved.

In order to calculate the entire NMR line shape, not just its second moment, one needs to know explicitly the probability distribution $W(p_1,p_2,p_3)$ as a function of local strains p_1,p_2,p_3. Here we only present a formal result, which can be derived from the definition (38):

$$W(p_1,p_2,p_3) = |\mathcal{J}|\exp\left(-\frac{1}{2}\sum_\mu x_\mu^2(p_1,p_2,p_3)\right) \ , \quad (44)$$

where each of the three fields $x_\mu(p_1,p_2,p_3)$ is to be calculated for a given set of values of local strains p_μ from the three coupled equations (29), and $\mathcal{J}$ is the Jacobian of the transformation. We find using Eqs. (29)

$$\mathcal{J} = \left(\frac{1}{\beta J\sqrt{q+\tilde{\Delta}}}\right)^3\frac{(1-t_1t_2t_3)^6}{(1-t_1^2)(1-t_2^2)(1-t_3^2)}\left[\frac{1}{(1-t_1^2t_2^2)(1-t_1^2t_3^2)(1-t_2^2t_3^2)}+\frac{2}{t_1t_2t_3(t_1^2-1)(t_2^2-1)(t_3^2-1)}\right.$$
$$\left.-\frac{1}{t_1^2(t_2^2-1)(t_3^2-1)(1-t_2^2t_3^2)}-\frac{1}{t_2^2(t_1^2-1)(t_3^2-1)(1-t_1^2t_3^2)}-\frac{1}{t_3^2(t_2^2-1)(t_1^2-1)(1-t_2^2t_1^2)}\right] , \quad (45)$$

where $t_\mu \equiv \tanh(\beta H_\mu)$, and H_μ is again given by Eq. (25).

IV. STABILITY OF THE ISOTROPIC SYMMETRIC PHASE

So far we have considered the equilibrium properties of the replica-symmetric quadrupolar glass phase without specifying the conditions for its stability. As shown in Sec. III, the replica-symmetric solution for the order parameter q_μ turns out to be isotropic with respect to the symmetry indices μ if no symmetry-breaking external fields are present. Following the well-known arguments from the theory of spin glasses,[17] however, one may expect that on lowering the temperature an instability with respect to replica-symmetry-breaking modes will appear.

As usual, we consider Gaussian fluctuations arround the isotropic replica-symmetric solution (28) and write

$$q_{\mu\nu}^{\alpha\beta} = q + \eta_\mu^{\alpha\beta} \ , \quad (46)$$

where we allow for the possibility that the fluctuations $\eta_\mu^{\alpha\beta}$ may depend on the symmetry indices μ. Here we assume that $J_0' = 0$ or equivalently $P_\mu = 0$. Without any loss of generality we may require[18]

$$\sum_{\beta} \eta_{\mu}^{\alpha\beta} = 0 \,. \tag{47}$$

Inserting the ansatz (46) into expression (22) and expanding up to second order in the small fluctuations $\eta_{\mu}^{\alpha\beta}$, we find the deviation of the free energy δf from the saddle-point value (24). This deviation can be expressed in terms of the appropriate averages of Z_{μ} variables as follows:

$$\delta f = -\frac{1}{4n}\beta^2 J^2 \Bigg[\sum_{\alpha\neq\beta} \sum_{\mu} \left(\eta_{\mu}^{\alpha\beta}\right)^2 - 2 \sum_{\alpha\neq\beta,\gamma\neq\delta} \sum_{\mu\nu} \Big(\langle Z_{\mu}^{\alpha} Z_{\mu}^{\beta} Z_{\nu}^{\gamma} Z_{\nu}^{\delta} \rangle - \langle Z_{\mu}^{\alpha} Z_{\mu}^{\beta} \rangle \langle Z_{\nu}^{\gamma} Z_{\nu}^{\delta} \rangle \Big) \eta_{\mu}^{\alpha\beta} \eta_{\nu}^{\gamma\delta} \Bigg] \,. \tag{48}$$

The expression in the square brackets has the following general structure:

$$\delta f = \sum_{\mu\nu} \sum_{\alpha\beta} (a_0^{\mu\nu} + a_1^{\mu\nu} + 2a_2^{\mu\nu}) \eta_{\mu}^{\alpha\beta} \eta_{\nu}^{\alpha\beta} (1 - \delta_{\alpha\beta}) + \sum_{\mu\nu} \sum_{\alpha\beta\gamma} (a_1^{\mu\nu} + 4a_2^{\mu\nu}) \eta_{\mu}^{\alpha\beta} \eta_{\nu}^{\beta\gamma} (1 - \delta_{\alpha\beta})(1 - \delta_{\beta\gamma})(1 - \delta_{\alpha\gamma}) + \sum_{\mu\nu} \sum_{\alpha\beta\gamma\delta} a_2^{\mu\nu} \eta_{\mu}^{\alpha\beta} \eta_{\nu}^{\gamma\delta} (1 - \delta_{\alpha\beta})(1 - \delta_{\gamma\delta})(1 - \delta_{\alpha\gamma})(1 - \delta_{\beta\delta}) \,, \tag{49}$$

where the diagonal $a_i^{\mu\mu}$ and off-diagonal $a_i^{\mu\nu}$ $(\mu \neq \nu)$ coefficients can be determined by a comparison between the two expressions (49) and (48). By evaluating the averages of Z_{μ} variables at the saddle point, we find

$$a_0^{\mu\mu} = 1 - \beta^2 J^2 \left(1 - 2q + \left[p_{\mu}^4\right]_{123}\right) \tag{50}$$

and

$$a_0^{12} = -\beta^2 J^2 \left(q + \left[p_1^2 p_2^2\right]_{123}\right) \,, \tag{51}$$

as well as two analogous equations for a_0^{13} and a_0^{23}. Here p_{μ}, $\mu = 1, 2, 3$, are local polarizations introduced in Eq. (29), and $q = q_{\mu}$ is the replica-symmetric order parameter given by Eq. (28). The expressions for the remaining coefficients $a_1^{\mu\nu}$ and $a_2^{\mu\nu}$ can also be found; however, due to the constraint (47) they do not enter explicitly the stability criterion, which now reduces to the following condition:

$$\delta f = \sum_{\mu\nu} \sum_{\alpha\neq\beta} \left[a_0^{\mu\mu} \eta_{\mu}^{\alpha\beta} \eta_{\mu}^{\alpha\beta} \delta_{\mu\nu} + a_0^{\mu\nu} \eta_{\mu}^{\alpha\beta} \eta_{\mu}^{\alpha\beta} (1 - \delta_{\mu\nu}) \right] \geq 0 \,. \tag{52}$$

From the expressions (50) and (51) and the corresponding expressions for local polarizations (29) and order parameters q_{μ} (28), we see that all diagonal elements $a_0^{\mu\mu}$ are equal, i.e., $a_0^{\mu\mu} = a_0^{11}$, and similarly all the off-diagonal elements are $a_0^{\mu\nu} = a_0^{12}$. Having this in mind, we realize that the condition (52) implies the positivity of the nondegenerate eigenvalue λ_1 as well as the doubly degenerate eigenvalues λ_2, which are given by

$$\lambda_1 = a_0^{11} + 2a_0^{12} \,, \tag{53}$$

$$\lambda_2 = a_0^{11} - a_0^{12} \,. \tag{54}$$

The corresponding eigenvectors are

$$\eta_1 = \begin{pmatrix} 1 \\ 1 \\ 1 \end{pmatrix} , \tag{55}$$

belonging to λ_1, while in the case of λ_2 one has

$$\eta_2 = \begin{pmatrix} 1 \\ -1 \\ 0 \end{pmatrix} , \tag{56}$$

$$\eta_2' = \begin{pmatrix} 1 \\ 0 \\ -1 \end{pmatrix} . \tag{57}$$

Therefore, the stability of the isotropic replica-symmetric solution (28) is violated if one of the eigenvalues (53) or (54) becomes zero. In terms of the more familiar random averages the two limits of stability can be expressed as

$$1 - \beta^2 J^2 \left(1 + \left[p_1^4 + 2p_1^2 p_2^2\right]_{123}\right) = 0 \,, \tag{58}$$

$$1 - \beta^2 J^2 \left(1 - 3q + \left[p_1^4 - p_1^2 p_2^2\right]_{123}\right) = 0 \,. \tag{59}$$

By solving numerically each of these equations together with Eq. (28) for the order parameter q, we find that the condition (58) is satisfied at a higher temperature $T_Q(\Delta)$ than the condition (59) for all values of Δ. Therefore, Eq. (58) determines the instability line in our model. In Fig. 4 the instability line $T_Q(\Delta)$ is plotted against the random-field variance $\tilde{\Delta}$. For comparison, the instability line $T_I(\Delta)$ for dipolar glasses obtained from the RB-RF Ising model[6] using the same parameter values is also shown. It turns out that the instability temperature of the discrete-state quadrupolar glass $T_Q(\Delta)$ always lies above the one obtained for the RB-RF Ising model[6] at the same value of $\tilde{\Delta}$.

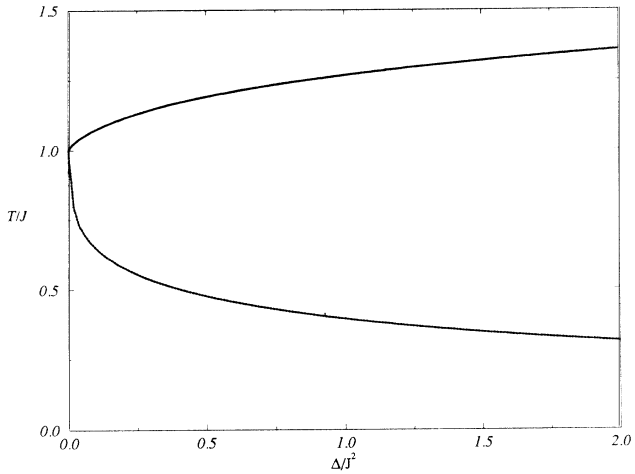

FIG. 4. Instability temperature T_Q/J of the isotropic replica-symmetric phase plotted vs reduced random-field variance Δ/J^2 (solid line). Also plotted is the instability temperature for the RB-RF Ising model (dotted line).

V. DISCUSSION AND CONCLUSIONS

The model of interacting quadrupoles presented in this paper is characterized by three essential features: (i) Due to a strong hindering potential the quadrupolar degrees of freedom are described by a set of discrete states corresponding to the equilibrium orientations along the $\langle 111 \rangle$ cubic directions; (ii) quenched shear strains—which transform according to the T_{2g} representation of the cubic group—give rise to quenched local random fields which couple to the symmetry-adapted projection operators $Z_{i\mu}$ describing the quadrupolar orientations; (iii) also included are infinite-range quenched random interactions between pairs of $Z_{i\mu}$ variables. This model is expected to be applicable to the orientational-glass phase in $K(CN)_xBr_{1-x}$ and similar mixed cyanide crystals for the range of concentrations of CN^- ions x below $x_c \approx 0.7$, where long-range order disappears. In our model, both the variance Δ of the random-field components and the variance J of the infinite-range random interactions are concentration dependent, and appear as phenomenological parameters in the theory.

Applying the replica mean-field theory we have derived the temperature dependence of the orientational-glass order parameter q, the linear susceptibility χ, and the shear elastic constant C_{44} in the isotropic replica-symmetric orientational-glass phase. We have also determined the instability temperature $T_Q(\Delta)$, below which at least one of the components of the order-parameter field q_μ becomes unstable against replica-symmetry-breaking fluctuations. The physical meaning of this instability relates to the fact that the response of the system to an applied external field starts to depend on the history of the system; i.e., field-cooled χ_{FC} and zero-field-cooled χ_{ZFC} susceptibilities, which are equal for temperatures T above $T_Q(\Delta)$, begin to differ for $T \leq T_Q(\Delta)$. Thus the instability temperature in the presence of random fields, $T_Q(\Delta)$, may be identified as the true freezing temperature. The splitting between χ_{FC} and χ_{ZFC} observed both in dipolar[19] and quadrupolar[9] glasses suggests that the random interaction plays an important role in these systems. In contrast to other models of quadrupolar glasses,[8,12,13] we require the presence of both random interactions and random fields in order to describe properly the experimental data obtained in these systems. Namely, models with random fields alone and no random interactions would not be capable of describing the experimentally observed splitting between χ_{FC} and χ_{ZFC} and the divergence of the nonlinear susceptibility χ_{NL}. On the other hand, models with random interactions alone and no random fields correctly describe the cooperative ordering, which is responsible for the freezing transition into a quadrupolar glass phase; however, they imply $q(T) = 0$ above the transition temperature and hence cannot be directly applied to the present system, where $q(T)$ has an experimentally observable high-temperature tail.

One should perhaps redefine the order parameter for systems containing both random bonds and random fields by considering the difference between the appropriate stable solution in each temperature range and the replica-symmetric solution $q(T)$—a point which deserves further attention. By construction, such an order parameter would measure just the cooperative glassy ordering and would in general vanish above $T_Q(\Delta)$. It is common, however, to refer to $q(T)$ as the quadrupolar glass order parameter, since it represents an experimentally accessible quantity[15] analogous to the magnetic-field-induced magnetization in the paramagnetic phase of a ferromagnet. Strictly speaking, the high-temperature phase cannot be regarded as a true quadrupolar glass—in spite of $q(T)$ being nonzero—but should rather be referred to as the random-field-induced Edwards-Anderson[21] order.

It is interesting to mention that the instability temperature $T_Q(\Delta)$ in our model appears to be slightly above the maximum of the linear susceptibility, in qualitative agreement with the experimental results of Hessinger and Knorr.[9] It has recently been shown[20] using a dynamic theory based on the soft-spin RB-RF Ising model that the nonlinear susceptibility χ_{NL} of dipolar glasses diverges at the instability line $T_I(\Delta)$, i.e., at the same temperature where the splitting between the field-cooled χ_{FC} and zero-field-cooled χ_{ZFC} linear susceptibilities occurs. Experiments on the quadrupolar glass $K(CN)_xBr_{1-x}$ (Ref. 9) show, however, that the divergence of χ_{NL} occurs close to, but not at the same temperature as the splitting between χ_{FC} and χ_{ZFC}. A theoretical explanation of this effect can only be sought within the framework of the appropriate dynamical model, and will be left for future work.

We have also derived an expression for the probability distribution of local strains and the NMR line shape, the second moment of which is directly proportional to the orientational-glass order parameter q. ^{14}N NMR second-moment measurements can be thus used for a direct determination of the glassy order parameter in quadrupolar glasses such as $K(CN)_xBr_{1-x}$.

In the $\Delta = 0$ plane, according to our model the ferroelastic phase becomes inevitable at low temperatures,

similar to the case of continuous model studied in Ref. 12. In the present case, the condition that the orientational-glass transition temperature is higher than the one of the ferroelastic phase transition becomes $J_0/J \leq 0$; i.e., the average random interaction must be kept nonpositive. The presence of random fields ($\Delta \neq 0$) is expected to reduce the region of parameter space where long-range order occurs, thus leading to a possible relaxation of the above condition. A schematic phase diagram in the presence of random fields is shown in Fig. 5. The appearance of long-range ferroelastic order in the range of concentrations of CN^- ions $x \leq 0.7$ might be an artifact of the mean-field theory. In reality, however, the formation of microdomains due to both random interactions and random fields prevents the onset of long-range order in this range of concentrations.

In the present model, only the shear strains are coupled to the components of the order-parameter field, and thus only elastic modulus C_{44} exhibits an anomalous temperature dependence due to orientational-glass ordering. Theoretical predictions of $C_{44}(T)$, given by Eq. (37), involve the phenomenological coupling parameter $\tilde{\gamma}$, which may be also concentration dependent.[3]

Below the instability temperature $T_Q(\Delta)$, replica-symmetry breaking presumably occurs, which may also include breaking of the isotropy between the components of the orientational-glass order parameter matrix q_μ. Theoretical investigations of the replica-symmetry breaking and its consequences on the relevant physical properties of the system is deferred for a future study.

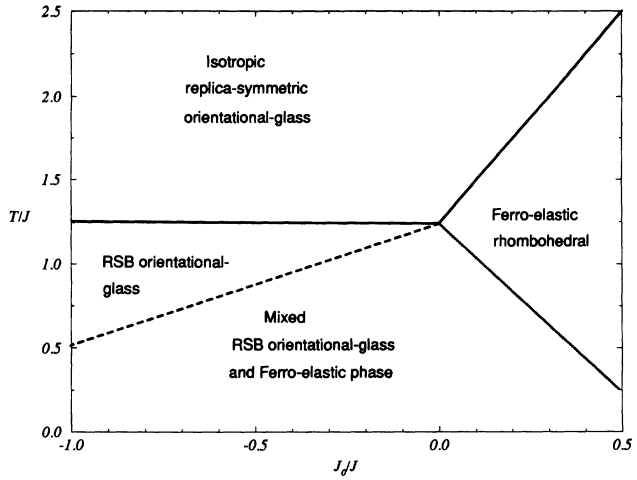

FIG. 5. Schematic phase diagram in the (T, J_0) plane for nonvanishing random-field variance $\tilde{\Delta} \neq 0$.

ACKNOWLEDGMENTS

This work was supported by the Ministry of Science and Technology of the Republic of Slovenia and by the Bundesministerium für Forschung und Technologie of the Federal Republic of Germany. Two of us (B.T. and R.P.) wish to acknowledge helpful discussions with Professor K. Knorr, Dr. J.K. Krüger, and Dr. R. Jiménez.

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