Dynamics of quadrupolar glasses

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The dynamic properties of quadrupolar glasses are investigated on the basis of a soft-spin version of the recently introduced symmetry adapted random-bond–random-field model. The model is applicable to the mixed alkali cyanides and related cubic systems with ⟨100⟩, ⟨111⟩, and ⟨110⟩ equilibrium orientations of the quadrupolar axis. Following the dynamic theories of Ising and vector spin glasses, a field-theoretic approach is formulated based on the Langevin equation of motion for the symmetry adapted order parameter fields. It is shown that the average relaxation time diverges on the line of instability $T_f(h_r)$, where T_f is the freezing temperature and h_r the strength of the static random strain fields, in agreement with the static replica theory. For the ⟨111⟩ model the long-time correlation function exponent ν is evaluated along the instability line. [S0163-1829(96)00534-6]

I. INTRODUCTION

Recently, the symmetry adapted random-bond–random-field (SARBRF) model of quadrupolar glasses (QG's) was introduced and its static properties were investigated.[1,2] The essential idea of the SARBRF model is that the order parameter fields should be defined in terms of symmetrized linear combinations of the discrete-state occupation number operators introduced earlier by Vollmayr *et al.*[3] In systems with strong cubic anisotropy the orientational degrees of freedom are described by a set of $s=3$, 4, or 6 discrete states corresponding to the ⟨100⟩, ⟨111⟩, or ⟨110⟩ equilibrium orientations, respectively, of the quadrupolar axis. A physical example are the mixed alkali cyanide-halide systems $KBr_{1-x}(CN)_x$, $K_{1-x}Na_xCN$ and related compounds,[4] where the anisotropy of the rotational potential for the CN^- ions is of the order ~ 35 K and is thus relevant at temperatures near the freezing transition.

Several other static theories of QG's have been formulated so far.[3,5,6] Specifically, in Ref. 3 it was shown that the discrete-state model for strongly anisotropic systems is equivalent to an s-state Potts glass.[7] In contrast to magnetic systems, local random strain fields are linearly coupled to the order parameter and hence need to be included into the model. Since these transform according to the irreducible representations of the local, i.e., cubic symmetry group of the pure system, the symmetrized linear combinations of the discrete-state occupation numbers appear as a natural choice for the order parameter fields. The resulting SARBRF model with $r \equiv s-1$ relevant order parameter fields and an isotropic random-bond interaction has so far been used within a replica approach to calculate the temperature dependence of the QG order parameter as well as the line of instability separating the ergodic from the nonergodic QG phase.[1,2]

For the limiting case of weak anisotropy potential, a natural choice for the symmetry adapted order parameter fields is a set of cubic harmonics $Y_\lambda(\vartheta,\varphi)$, $\lambda = 1,2,\ldots,5$,[8] where the angular variables specify the molecular orientations.

As far as dynamic theories are concerned, the soft-spin Potts glass model without random fields has been investigated by Thirumalai and Kirkpatrick.[9] The problem of dielectric relaxation in quadrupolar glasses has been studied by Kanter and Sompolinsky[10] using a generalized randomly anisotropic quadrupolar interaction. In the present paper we will adopt an analogous form for the random interaction within the SARBRF model. We will focus on the dynamics of this model on and above the line of instability, following the well-known examples of the soft-spin dynamics of magnetic spin glasses[11–13] and/or dipolar glasses described by the Ising random-bond–random-field (RBRF) model.[14] A transformation to continuous order parameter fields will be carried out via field-theoretic methods, i.e., by introducing the appropriate field densities, which contain all the symmetries of the static SARBRF model.

In Sec. II we introduce the SARBRF model in its general form and write down the corresponding Langevin equations of motion. The averaged local equation of motion is derived in Sec. III and an effective relaxation rate is introduced. In Sec. IV the line of instability is derived and evaluated for three representative cases. The long-time scaling exponent ν is calculated in Sec. V for the special case of the ⟨111⟩ model. Finally, in Sec. VI the conclusions are presented.

II. DYNAMIC SARBRF MODEL

The Hamiltonian of the soft-spin SARBRF model is formally written as

$$\mathcal{H} = -\frac{1}{2}\sum_{ij}\sum_{\mu,\nu=1}^{r} J_{ij}^{\mu\nu}\phi_{i\mu}\phi_{j\nu} - \sum_i \sum_{\mu=1}^{r} (h_{i\mu}+E_{i\mu})\phi_{i\mu} + \frac{1}{\beta}\sum_i \rho[\phi_i], \tag{1}$$

where $\mu,\nu = 1,2,\ldots,r$ with $r \equiv s-1$ label the symmetry components of the order parameter fields ϕ_i, local random strain fields h_i, and external fields E_i. We assume a general anisotropic form for the random-bond interaction $J_{ij}^{\mu\nu}$.

In the static SARBRF model, the fields $\phi_{i\mu}$ are replaced by discrete variables $Z_{i\mu}$, which are defined as linear combinations of the occupation number operators $N_{ip}=0,1$ for each set of discrete orientations $p=1,2,\ldots,s$, namely,

$$Z_{i\mu}=\sum_{p=1}^{s} a_{\mu p}N_{ip}. \tag{2}$$

The coefficients $a_{\mu p}$ are simply determined by group theory arguments. Equation (2) includes the trivial case $Z_{is}=1$, with $a_{sp}=1$ in view of the closure relation $\Sigma_p N_{ip}=1$. Dropping the site index i, the relevant order parameter fields are explicitly

$$\langle 100\rangle: \quad Z_1=\sqrt{3/2}(N_1-N_2), \quad Z_2=\sqrt{1/2}(2N_3-N_1-N_2). \tag{3}$$

$$\begin{aligned}\langle 111\rangle: \quad Z_1&=N_1+N_2-N_3-N_4,\\ Z_2&=N_2+N_3-N_1-N_4,\\ Z_3&=N_3+N_1-N_2-N_4,\end{aligned} \tag{4}$$

$$\begin{aligned}\langle 110\rangle: \quad Z_1&=\sqrt{3/2}(N_2+N_5-N_3-N_6),\\ Z_2&=\sqrt{1/2}(2N_1+2N_4-N_2-N_3-N_5-N_6),\\ Z_3&=\sqrt{3}(N_1-N_4), \quad Z_4=\sqrt{3}(N_2-N_5),\\ Z_5&=\sqrt{3}(N_3-N_6).\end{aligned} \tag{5}$$

The fields Z_1 and Z_2 in Eqs. (3) and (5) transform according to the E_g representation of the cubic group, whereas the remaining triplet fields transform according to T_{2g}.

The last term in Eq. (1) represents the local order-parameter field density, which can be derived by the usual field-theoretic methods (see the Appendix). Its general form for the present model is

$$\rho[\phi]=\frac{1}{2}r_0\sum_{\mu}\phi_\mu^2+\frac{1}{3}\sum_{\mu\nu\kappa}w_{\mu\nu\kappa}\phi_\mu\phi_\nu\phi_\kappa + \frac{1}{4}\sum_{\mu\nu\kappa}u_{\mu\nu\kappa}\phi_\mu\phi_\nu\phi_\kappa^2+\cdots. \tag{6}$$

The parameters r_0, $w_{\mu\nu\kappa}$, $u_{\mu\nu\kappa}$, etc., are fixed by the requirement that the soft-operator representation should reproduce the known static properties of the model. Symmetry restricts the coefficients $w_{\mu\nu\kappa}$, $u_{\mu\nu\kappa}$ to a set of nonzero values in each particular case, the simplest being the $\langle 111\rangle$ model where

$$\begin{aligned}w_{\mu\nu\kappa}&=w \quad \text{for} \quad \mu\neq\nu\neq\kappa,\\ u_{\mu\nu\kappa}&=u \quad \text{for} \quad \mu=\nu=\kappa,\end{aligned} \tag{7}$$

and $w_{\mu\nu\kappa}=u_{\mu\nu\kappa}=0$ otherwise.

Finally, the quenched infinite range random interactions $J_{ij}^{\mu\nu}$ and random fields $h_{i\mu}$ in Eq. (1) are assumed to be uncorrelated and characterized by their respective Gaussian distributions, i.e.,

$$[J_{ij}^{\mu\nu}]_{\rm av}=0, \quad [(J_{ij}^{\mu\nu})^2]_{\rm av}=J^2/(Nr), \tag{8}$$

and

$$[h_{i\mu}]_{\rm av}=0, \quad [h_{i\mu}h_{j\nu}]_{\rm av}=\delta_{ij}\delta_{\mu\nu}h_r^2J^2/r. \tag{9}$$

The randomly anisotropic interaction $J_{ij}^{\mu\nu}$ (Refs. 8 and 10) of the above type is believed to be physically more appropriate than the isotropic scalar interaction $J_{ij}^{\mu\nu}=J_{ij}\delta_{\mu\nu}$ frequently used in static theories.[1,2] In particular, the random average of type (8) has the advantage that it does not lead to long-range ferroelastic order at any temperature. By contrast, in the scalar model one has to assume a nonzero average $[J_{ij}]_{\rm av}=J_0/N<0$ in order to eliminate the spontaneous strains at least in a certain temperature range.[1,2]

Following the dynamic theories of spin glasses[15] we assume that the time dependent order parameter fields $\phi_{i\mu}(t)$ obey the Langevin equations of motion

$$\Gamma_0^{-1}\frac{\partial}{\partial t}\phi_{i\mu}=-\frac{\partial(\beta\mathcal{H})}{\partial\phi_{i\mu}}+\xi_{i\mu}, \tag{10}$$

where Γ_0 is the bare kinetic coefficient, and $\xi_{i\mu}$ the Gaussian stochastic noise with zero average and correlations

$$\langle\xi_{i\mu}(t)\xi_{j\nu}(t')\rangle=\frac{2}{\Gamma_0}\delta_{ij}\delta_{\mu\nu}\delta(t-t'). \tag{11}$$

We will consider the correlation and response functions[11,14]

$$C_{\mu\nu}(t-t')=\frac{1}{N}\sum_i\langle\phi_{i\mu}(t)\phi_{i\nu}(t')\rangle_\xi, \tag{12}$$

$$G_{\mu\nu}(t-t')=\frac{1}{N}\sum_i\frac{\partial\langle\phi_{i\mu}(t)\rangle_\xi}{\partial E_{i\nu}(t')}, \quad t>t'. \tag{13}$$

The symbol $\langle\cdots\rangle_\xi$ implies averaging over the Gaussian noise ξ.

III. EFFECTIVE RELAXATION RATE

It is by now standard in spin glass theory[11,15] to apply the functional integral formalism to the dynamics of the order parameter fields, thus enabling one to perform the averages over the probability distributions of random bonds and random fields without the use of replicas. Here we will not describe the details of this formulation, which is similar to the case of vector spin glasses.[12,13] It should be noted, however, that the algebra of the ϕ_μ operators differs from that of vector spins, and there are new features introduced by the cubic $w_{\mu\nu\kappa}$ terms in the field density (6).

We proceed to the effective equation of motion for the Fourier components $\phi_\mu(\omega)$ of the order parameter fields,

$$\sum_{\nu}\,[\mathbf{G}^{0}(\omega)^{-1}]_{\mu\nu}\phi_{\nu}(\omega)=\psi_{\mu}(\omega)+\beta E_{\mu}(\omega)-\sum_{\nu\kappa}\,w_{\mu\nu\kappa}\int\frac{d\omega_1}{2\pi}\phi_{\nu}(\omega_1)\phi_{\kappa}(\omega-\omega_1)-\frac{1}{2}\sum_{\nu\kappa}\,u_{\mu\nu\kappa}\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\phi_{\nu}(\omega_1)$$
$$\times[\phi_{\mu}(\omega_2)+\phi_{\kappa}(\omega_2)]\phi_{\kappa}(\omega-\omega_1-\omega_2), \tag{14}$$

where the bare response $\mathbf{G}^{0}(\omega)$ is an $r\times r$ matrix, which satisfies the equation

$$\mathbf{G}^{0}(\omega)^{-1}=(r_0-i\omega/\Gamma_0)\mathbf{1}-\beta^2J^2\mathbf{G}(\omega). \tag{15}$$

The matrix elements of the full propagator $\mathbf{G}(\omega)$ are defined by Eq. (13). It will be argued below that the response is diagonal, i.e., $G_{\mu\nu}(\omega)=\delta_{\mu\nu}G(\omega)$. Therefore, the off-diagonal part of $\mathbf{G}^{0}(\omega)^{-1}$ in Eq. (14) is identically zero, but is formally included in order to preserve the general structure of the equation of motion.

In Eq. (14) $\psi_{\mu}(\omega)$ is the Fourier component of an effective stochastic noise $\psi_{i\mu}(t)$, which can be decomposed into a fluctuating part $f_{i\mu}(t)$ and an excess static noise $x_{i\mu}$, i.e.,

$$\psi_{i\mu}(t)=f_{i\mu}(t)+x_{i\mu}. \tag{16}$$

The corresponding correlations are

$$\langle x_{i\mu}x_{j\nu}\rangle=\delta_{ij}\delta_{\mu\nu}(q+h_r^2), \tag{17}$$

$$\langle f_{i\mu}(t)f_{j\nu}(t')\rangle=\delta_{ij}[2\Gamma_0^{-1}\delta_{\mu\nu}\delta(t-t')+\beta^2J^2\widetilde{C}_{\mu\nu}(t-t')], \tag{18}$$

where $\widetilde{C}_{\mu\nu}(t-t')\equiv C(t-t')_{\mu\nu}-q_{\mu\nu}\delta_{\mu\nu}$ is the fluctuating part of the correlation function (12), which has a nonzero value in the long-time limit, namely,

$$\lim_{(t-t')\to\infty}\,C_{\mu\nu}(t-t')=q\,\delta_{\mu\nu}. \tag{19}$$

The quantity $q_{\mu\nu}$, which plays the role of QG order parameter matrix known from the static theories,[1,2] and has nonzero components at any temperature due to random strains.

In view of the decomposition (16) it is convenient to investigate the response at a fixed value of the static noise field $\vec{x}=(x_1,x_2,\ldots,x_r)$, which will be denoted by $\mathbf{g}(\omega,\vec{x})$. This is related to the response $\mathbf{G}(\omega)$ according to

$$[g_{\mu\nu}(\omega,\vec{x})]_{\vec{x}}=G(\omega)\,\delta_{\mu\nu}, \tag{20}$$

where we make use of the fact that averaging has restored the global cubic symmetry.

The function $\mathbf{g}(\omega,\vec{x})$ satisfies the Dyson equation

$$\mathbf{g}^{-1}(\omega,\vec{x})=\mathbf{G}^{0}(\omega)^{-1}+\mathbf{\Sigma}(\omega,\vec{x}), \tag{21}$$

where $\mathbf{\Sigma}(\omega,\vec{x})$ represents the self-energy, which is determined by the $w_{\mu\nu\kappa}$ and $u_{\mu\nu\kappa}$ terms in Eq. (14). The symbol $[\cdots]_{\vec{x}}$ means a Gaussian average over the static noise.

We now define an effective kinetic coefficient $\hat{\Gamma}$ through the relation

$$\hat{\Gamma}^{-1}=-i\lim_{\omega\to0}\left(\frac{\partial}{\partial\omega}G(\omega)\right). \tag{22}$$

From Eqs. (15) and (21) we then have

$$\mathbf{g}^{-1}(\omega,\vec{x})=(r_0-i\omega/\Gamma_0)\mathbf{1}-\beta^2J^2\mathbf{G}(\boldsymbol{\omega})+\mathbf{\Sigma}(\omega,\vec{x}). \tag{23}$$

Differentiating with respect to ω, multiplying by $\mathbf{g}(\omega,\vec{x})$ from each side, and averaging over $\vec{x}$ we obtain

$$(\mathbf{1}-\beta^2J^2[\mathbf{g}^2]_{\vec{x}})\hat{\Gamma}^{-1}=\Gamma_0^{-1}[\mathbf{g}^2]_{\vec{x}}+i[\mathbf{g}(\partial\mathbf{\Sigma}/\partial\omega)\mathbf{g}]_{\vec{x}}, \tag{24}$$

where $\mathbf{g}\equiv\mathbf{g}(0,\vec{x})$ and the limit $\omega\to0$ is understood.

The quantity $g_{\mu\nu}(0,\vec{x})$ is related to the static susceptibility $\chi_{\mu\nu}(\vec{x})$ in a fixed field $\vec{x}$:

$$\chi_{\mu\nu}(\vec{x})=\beta g_{\mu\nu}(0,\vec{x}). \tag{25}$$

IV. STATIC LIMIT AND THE LINE OF INSTABILITY

In the static limit, $g_{\mu\nu}(0,\vec{x})$ as calculated from Eq. (23) should agree with the result obtained from the static theory based on the discrete fields $Z_{i\mu}$, by analogy to the Ising case.[11,14,15] Thus one can write

$$g_{\mu\nu}=\langle Z_{\mu}Z_{\nu}\rangle-p_{\mu}p_{\nu}, \tag{26}$$

where $p_{\mu}=\langle Z_{\mu}\rangle$ and the thermodynamic average is evaluated with the effective static Hamiltonian

$$\mathcal{H}_{\rm eff}=J\sqrt{q+h_r^2}\sum_{\mu}\,x_{\mu}Z_{\mu}. \tag{27}$$

Here q is a scalar QG order parameter obtained from the static replica theory. It should be noted that in the static SARBRF model, the order parameter matrix $q_{\mu\nu}$ is diagonal in the high-temperature phase, namely,

$$q_{\mu\nu}=[p_{\mu}p_{\nu}]_{\vec{x}}=q\,\delta_{\mu\nu}. \tag{28}$$

Thus the diagonal components $q_{\mu\mu}$ are given in terms of a single longitudinal parameter $q_L=q$, whereas the transverse components, $q_T=q_{\mu\nu}$ $(\mu\neq\nu)$, are strictly zero. This can be easily verified numerically for the three models $\langle100\rangle$, $\langle111\rangle$, and $\langle110\rangle$ as discussed in more detail in Refs. 1 and 2. The above simple parametrization of the order parameter is inherent to the SARBRF model, and implies that, on the average, the system is invariant under transformations of the cubic group.

In a similar fashion one can explicitly prove the relation

$$\sum_{\nu}\,[g_{\nu\kappa}(0,\vec{x})g_{\mu\nu}(0,\vec{x})]_{\vec{x}}=\delta_{\mu\kappa}[(\mathbf{g}^2(0,\vec{x}))_{\mu\mu}]_{\vec{x}}, \tag{29}$$

TABLE I. Values of the constant b from Eqs. (32) and (33) for various models, and line of instability $\theta \equiv \theta(h_r)$ in the limit of small random field $h_r \ll 1$.

Model	$\langle 100 \rangle$	$\langle 111 \rangle$	$\langle 110 \rangle$	Ising
b	1	2	4	0
θ	$h_r/\sqrt{2}$	$(7/2)h_r^2$	$h_r/\sqrt{2}$	$(3/4)^{1/3}h_r^{2/3}$

where the last average is, in fact, μ independent. Thus the matrices $[\mathbf{g}^2]_{\vec{x}}$ in Eq. (24) are diagonal and can be evaluated at any μ, say, $\mu=1$. For example, in the $\langle 111 \rangle$ case we find[16]

$$\sum_{\nu} [g_{\mu\nu}^2]_{\vec{x}} = \sum_{\nu} [g_{1\nu}^2]_{\vec{x}} = 1 + [p_1^4]_{\vec{x}} + 4[p_1 p_2 p_3]_{\vec{x}} + 2[p_1^2 p_2^2]_{\vec{x}}. \tag{30}$$

By analogy with dipolar glasses, the QG order parameter q is a measurable quantity and can be determined by NMR (Refs. 17 and 18) and related techniques.[19]

It can be shown that the term containing $\partial \mathbf{\Sigma}/\partial \omega$ in Eq. (24) remains finite as $\omega \to 0$. Therefore, one can solve Eq. (24) for $\hat{\Gamma}^{-1}$ provided that none of the eigenvalues of the matrix $\mathbf{1} - \beta^2 J^2 [\mathbf{g}^2]_{\vec{x}}$ is zero.[15] On the other hand, if one of its eigenvalues vanishes, this signals the occurrence of a dynamic instability. Combining Eqs. (24) and (25) we find that the instability will develop when

$$J^2 \left[\sum_{\nu} \chi_{\mu\nu}^2(\vec{x}) \right]_{\vec{x}} = 1. \tag{31}$$

Thus in the high-temperature phase the system will exhibit critical slowing down at the freezing temperature $T_f(h_r)$ obtained as the solution of Eq. (31). Formally, the same condition for $T_f(h_r)$ follows from the static replica theory if one uses either the randomly anisotropic interaction (8) or the scalar isotropic interaction $J_{ij}^{\mu\nu} = J_{ij}\delta_{\mu\nu}$.[2] The line of instability $T = T_f(h_r)$ separates the high-temperature noncooperative ergodic QG phase from the low-temperature cooperative nonergodic QG phase, and is analogous to the familiar AT line of spin glasses.[20] This analogy is meaningful provided the transition is continuous. However, in the s-state Potts glass without random fields ($h_r = 0$) the transition becomes discontinuous for $s > 4$ and the actual transition temperature lies above the zero-field value $T_f = J$.[21] Therefore, the result for the present $\langle 110 \rangle$ ($s=6$) case should be regarded as an estimated lower bound for the freezing temperature $T_f(h_r)$.

Close to T_f and for h_r small one can expand the averages in Eqs. (28) and (31) in powers of $\tilde{q} \equiv q + h_r^2$ and find to leading order

$$q = h_r^2 + \beta^2 J^2 \tilde{q} - \frac{1}{2}\beta^4 J^4 \tilde{q}^2 (4-b) + \mathcal{O}(\beta^6 J^6 \tilde{q}^3), \tag{32}$$

$$T^2/J^2 = 1 - \beta^2 J^2 \tilde{q}(2-b) + \mathcal{O}(\beta^4 J^4 \tilde{q}^2). \tag{33}$$

Here $b \equiv (1/r)\Sigma_{\mu\nu\kappa} \langle Z_\mu Z_\nu Z_\kappa \rangle_{h_r=0}^2 = r - 1$. The values of b for the various models are listed in Table I. In the $\langle 111 \rangle$ model one has $b=2$ and the second term in Eq. (33) vanishes, so that one has to evaluate the next term yielding

$$T_{\langle 111 \rangle}^2/J^2 = 1 - 7\beta^4 J^4 \tilde{q}^2 + \mathcal{O}(\beta^6 J^6 \tilde{q}^3). \tag{34}$$

In the $\langle 110 \rangle$ case, Eq. (33) becomes

$$T_{\langle 110 \rangle}^2/J^2 = 1 + 2\beta^2 J^2 \tilde{q} + \mathcal{O}(\beta^4 J^4 \tilde{q}^2), \tag{35}$$

indicating that T_f initially increases with $\tilde{q}$ and hence with h_r.

Introducing $\theta \equiv |T_f(h_r) - J|/J$, one can derive the line of instability for small h_r in analytic form $\theta = \theta(h_r)$. The results are summarized in Table I.

In Fig. 1 the numerical results for the instability line $T_f(h_r)$ in a broad temperature range for the $\langle 100 \rangle$, $\langle 111 \rangle$, and $\langle 110 \rangle$ models, i.e., $s=3$, 4, and 6, respectively, are shown together with the Ising RBRF case ($s=2$). As already mentioned, the $\langle 110 \rangle$ ($s=6$) case provides only an estimate of the actual line of instability as it would occur if the transition were continuous. The initial increase of T_f vs h_r may be related to this problem.

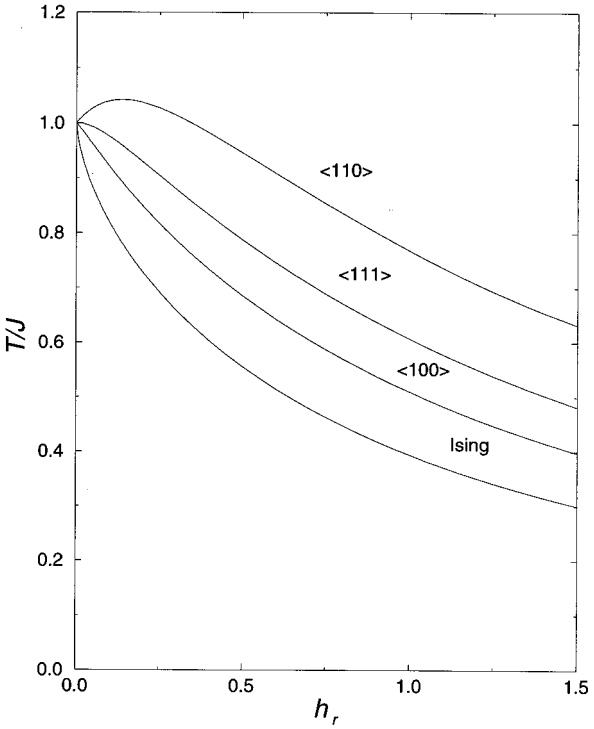

FIG. 1. Lines of dynamic instability $T = T_f(h_r)$ plotted vs random-field strength h_r, for quadrupolar glasses described by the SARBRF model with $\langle 100 \rangle$, $\langle 111 \rangle$, and $\langle 110 \rangle$ equilibrium orientations of the molecular axis. Also plotted is the instability line for dipolar glasses described by the Ising RBRF model.

One can define an effective relaxation time

$$\tau_{\text{eff}} = \frac{1}{q-1}\hat{\Gamma}^{-1}, \tag{36}$$

which diverges on approaching the line of instability from above. Introducing $\epsilon \equiv (T - T_f)/T_f$ as the vertical distance from a point $T_f = T_f(h_r)$ on this line, we find

$$\tau_{\rm eff} \propto \epsilon^{-\gamma}. \tag{37}$$

From Eqs. (24) and (31) one can see that the exponent γ takes the mean field value $\gamma=1$ and is independent of the strength of the random field h_r, i.e., γ is universal.

V. SLOW RELAXATION AT LONG TIMES

To characterize the dynamic properties of the model we focus on the long-time behavior of the correlation functions $\tilde{C}_{\mu\nu}(t)=\tilde{C}(t)\delta_{\mu\nu}$, where by analogy to the response $G(t)$ we need to consider only diagonal correlations. Anticipating a power-law asymptotic behavior at $t\to\infty$ as found in spin glasses and other glassy systems,[11,15] we introduce a scaling exponent ν by writing

$$\tilde{C}(t)\sim t^{-\nu}. \tag{38}$$

In the following we will limit ourselves to the $\langle 111\rangle$ model ($s=4$) in view of the particularly simple algebra of the corresponding Z_μ operators.[1] The calculation parallels that for the spin[15] and dipolar glasses,[14] however, there are new features in view of the presence of third-order coupling w. For the response function we use the ansatz[15]

$$\Delta G(\omega)\equiv\Delta G(\omega)-\Delta G(0)=-R(-i\omega/\Gamma)^{-\nu}, \tag{39}$$

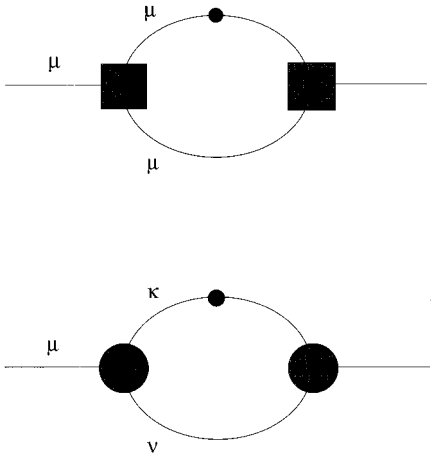

FIG. 2. Two types of diagrams for the derivative of self-energy $\partial\Sigma_\mu/\partial i\omega$ in the $\langle 111\rangle$ model. The line with a dot represents $\partial g_{\lambda\lambda}(\omega,\vec{x})/\partial i\omega$, where $\lambda=\mu$ or κ. Heavy squares and circles represent renormalized u and w vertices, respectively.

with Γ to be specified below. Following the examples from the theories of dipolar[14] and vector[13] glasses, we calculate the contributions of the self-energy diagrams in Fig. 2 with renormalized vertices and thus derive the relation

$$\Delta G(\omega)^2=2(A_u+A_w)\int_{-\infty}^{+\infty}\frac{d\omega'}{\pi\omega'}{\rm Im}\Delta G(\omega')[\Delta G(\omega+\omega')-\Delta G(\omega')]+\frac{i\omega}{\Gamma}, \tag{40}$$

where $\Gamma=\Gamma_0\beta^4J^4[\langle\mathbf{g}^3\rangle_0]_{\vec{x}}/[\langle\mathbf{g}^2\rangle_0]_{\vec{x}}$ and

$$A_u=[\langle(\delta Z_1)^3\rangle^2]_{\vec{x}}/[\langle\mathbf{g}^3\rangle_0]_{\vec{x}}, \tag{41a}$$

$$A_w=[\langle\delta Z_1\delta Z_2\delta Z_3\rangle^2]_{\vec{x}}/[\langle\mathbf{g}^3\rangle_0]_{\vec{x}}, \tag{41b}$$

with $\delta Z_\mu\equiv Z_\mu-p_\mu$ and $\langle\cdots\rangle_0\equiv{\rm Tr}(\cdots)/{\rm Tr}\mathbf{1}={\rm Tr}(\cdots)/r$. For the averages in Eqs. (41) we find

$$[\langle(\delta Z_1)^3\rangle^2]_{\vec{x}}=4[p_1^2(1-p_1^2)^2]_{\vec{x}}, \tag{42a}$$

$$[\langle\delta Z_1\delta Z_2\delta Z_3\rangle^2]_{\vec{x}}=[(1-p_1^2-p_2^2-p_3^2-2p_1p_2p_3)^2]_{\vec{x}}, \tag{42b}$$

$$[\langle\mathbf{g}^3\rangle_0]_{\vec{x}}=\left[\frac{1}{3}\sum_\mu g_{\mu\mu}^3+\sum_{\mu\nu}g_{\mu\mu}g_{\mu\nu}^2(1-\delta_{\mu\nu})+2g_{12}g_{23}g_{31}\right]_{\vec{x}}, \tag{42c}$$

where $g_{\mu\nu}$ are given by Eq. (26), which in the $\langle 111\rangle$ case becomes explicitly

$$g_{\mu\nu}=p_\kappa(\delta_{\mu\nu}-1)+\delta_{\mu\nu}-p_\mu p_\nu. \tag{43}$$

From Eq. (40) it follows that the value of the critical exponent ν on the instability line is determined by the equation

$$4\pi\cot(\pi\nu)=2B(\nu,\nu)(A_u+A_w), \tag{44}$$

where as usual[15] $B(\nu,\nu)$ is the β function. The solution of Eq. (44) representing the value of the long-time scaling exponent ν along the line of instability is shown in Fig. 3 together with the corresponding result for the Ising RBRF model. In the limit of zero random fields or $T/J\to1$ the exponent ν approaches the value $\nu=0.395$, which in the Ising case is found for $T=0$, and is here due to the fact that $A_w\to1/2$ in this limit, whereas $A_u\to0$. As the temperature is lowered, ν increases but remains $\nu<1/2$, and for $T\to0$ approaches the extrapolated value $\nu\simeq0.478$.

In the region below the instability line, the dynamics of glassy systems is characterized by a broad distribution of relaxation times resulting from a distribution $P(V)$ of potential barriers for quadrupolar reorientation. For a QG without local crystal-field anisotropy a calculation of $P(V)$ has been carried out in Ref. 10 using a mean field theory and the Parisi order parameter function $q(x)$.[15] An analogous calculation for the SARBRF model would be beyond the scope of the present paper, since the problem of replica-symmetry break-

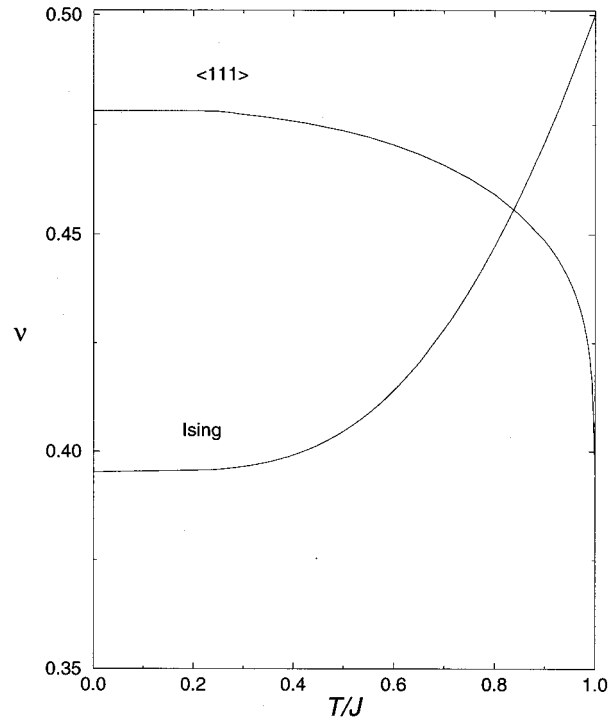

FIG. 3. Exponent ν plotted vs temperature along the line of instability $T=T_f(h_r)$ for a quadrupolar glass with $\langle 111\rangle$ equilibrium orientations. Also shown is the exponent ν for an Ising dipolar glass.

ing in the SARBRF model has not yet been solved. This will be the subject of a forthcoming publication.

VI. CONCLUSIONS

The main points of this paper are the following.

(1) A soft-spin symmetry adapted random-bond–random-field (SARBRF) model has been formulated to describe the dynamic properties of quadrupolar glasses (QG's) with specific reference to mixed crystals with cubic symmetry with $\langle 100\rangle$, $\langle 111\rangle$, or $\langle 110\rangle$ equilibrium orientations of the quadrupolar axis. For physical reasons the model includes a linear coupling between the order parameter field $\phi_{i\mu}$ and random local strain fields of strength h_r, which are represented by their irreducible components. An alternative description would be in terms of a soft-spin s-state Potts glass with random fields, where $s=3$, 4, or 6, respectively, for the above three cases. In the static SARBRF model, the order parameter field has only $r=s-1$ relevant irreducible components Z_μ, which are symmetrized linear combinations of the discrete-state occupation numbers. The random-bond interaction is assumed to have the general form $J_{ij}^{\mu\nu}$, where each component is an independent Gaussian random variable with zero mean, which ensures that there is no long-range order at any temperature.

(2) A soft-spin density $\rho[\phi]$ is constructed by using field-theoretic arguments, and dynamics is introduced via the Langevin equations for $\phi_{i\mu}$, from which an effective equation of motion for the soft-spin fields $\phi_\mu(\omega)$ is derived. The solutions are investigated in the region on and above the instability line $T_f(h_r)$, which separates the ergodic from the nonergodic QG phase. The freezing temperature T_f is calculated from the dynamic condition that the effective kinetic coefficient $\hat{\Gamma}^{-1}$ should diverge on the instability line in the limit $\omega\to 0$. The results for $T_f(h_r)$ agree with the static replica theory.

(3) The dynamic correlation function decays algebraically at long times with a characteristic exponent ν. An evaluation of ν along the instability line has been carried out for the case of a $\langle 111\rangle$ model. In contrast to spin and dipolar glasses, ν increases towards lower temperatures. This new feature is due to a third-order coupling in the field density $\rho[\phi]$.

(4) The freezing temperature $T_f(h_r)$ and the long-time exponent ν are observable quantities and could, in principle, be measured in mixed cyanides and related systems by observing the elastic response functions, for example, by ultrasonic methods, Brillouin scattering, and strain monitoring techniques.[19] In dipolar glasses, the exponent ν has been determined by dielectric spectroscopy,[22] however, no analogous experiments have been reported so far in quadrupolar glasses.

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APPENDIX: FIELD DENSITY

The partition function for a QG can be written with the aid of a Hubbard-Stratonovich transformation as a functional integral over the ''soft-spin'' fields $\phi_{i\mu}$:

$$\mathcal{Z}=\int \mathcal{D}\phi_{i\mu}\left\langle \exp\left(-\frac{1}{2}\beta^{-1}\sum_{ij}\sum_{\mu,\nu=1}^{r}(\mathbf{J}^{-1})_{ij}^{\mu\nu}\phi_{i\mu}\phi_{j\nu}+\sum_{i}\sum_{\mu=1}^{r}Z_{i\mu}\phi_{i\mu}\right)\right\rangle. \tag{A1}$$

Here $Z_{i\mu}$ are the discrete fields as defined by Eqs. (3)–(5) and $\langle\cdots\rangle\equiv \mathrm{Tr}(\cdots)/s$, where the trace involves a sum over all orientations. Again, in the exponent we ignore the trivial symmetry $\mu=s=A_{1g}$ for which $Z_{is}=1$. We introduce the density $\rho[\phi_i]$ by rewriting Eq. (A1) as

$$\mathcal{Z}=\int \mathcal{D}\phi_{i\mu}\exp\left(\sum_{i}\rho[\phi_i]\right)\exp\left(-\frac{1}{2}\beta^{-1}\sum_{ij}\sum_{\mu\nu}(\mathbf{J}^{-1})_{ij}^{\mu\nu}\phi_{i\mu}\phi_{j\mu}\right), \tag{A2}$$

implying

$$\rho[\phi_i]=\ln\left\langle \exp\left(\sum_{\mu} Z_{i\mu}\phi_{i\mu}\right)\right\rangle. \tag{A3}$$

Expanding up to fourth-order terms and dropping the site index i leads to

$$\rho[\phi]=\frac{1}{2}\sum_{\mu}\phi_{\mu}^2-\frac{1}{8}\left(\sum_{\mu}\phi_{\mu}^2\right)^2+\frac{1}{6}\sum_{\mu\nu\kappa}\langle Z_{\mu}Z_{\nu}Z_{\kappa}\rangle\phi_{\mu}\phi_{\nu}\phi_{\kappa}+\frac{1}{24}\sum_{\mu\nu\kappa\lambda}\langle Z_{\mu}Z_{\nu}Z_{\kappa}Z_{\lambda}\rangle\phi_{\mu}\phi_{\nu}\phi_{\kappa}\phi_{\lambda}+\cdots. \tag{A4}$$

Evaluating the averages by using the algebra of the discrete Z_{μ} operators for the three models as given by Eqs. (2)–(5), we realize that the general structure of the density $\rho[\phi]$ has the form given by Eq. (6). Considering the symmetry of the coefficients $w_{\mu\nu\kappa}$ and $u_{\mu\nu\kappa}$ in the three cases of interest, we obtain the following result:

$$\rho[\phi]_{\langle 100\rangle}=\frac{1}{2}(\phi_1^2+\phi_2^2)+u_1(\phi_1^4+\phi_2^4)+u_2\phi_1^2\phi_2^2+w_1\phi_1^2\phi_2+w_2\phi_2^3, \tag{A5}$$

$$\rho[\phi]_{\langle 111\rangle}=\frac{1}{2}(\phi_1^2+\phi_2^2+\phi_3^2)+\frac{1}{4}u(\phi_1^4+\phi_2^4+\phi_3^4)+w\phi_1\phi_2\phi_3, \tag{A6}$$

$$\rho[\phi]_{\langle 110\rangle}=\frac{1}{2}(\phi_1^2+\phi_2^2+\phi_3^2+\phi_4^2+\phi_5^2)+u_1(\phi_1^4+\phi_2^4)+\sum_{\mu\nu}u_{\mu\nu}\phi_{\mu}^2\phi_{\nu}^2+\sum_{\mu\nu}w_{\mu\nu}\phi_{\mu}\phi_{\nu}^2+w_1\phi_1^3+u_2\phi_1\phi_2(\phi_4^2-\phi_5^2). \tag{A7}$$

Comparing Eqs. (A4) and (A6) leads to relation (7).

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