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Dynamic approach to local-polarization distribution and NMR line shape in deuteron glasses

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The path-integral formulation of Glauber dynamics by Sommers is applied to calculate the deuteron NMR line shape in structural glasses such as $Rb_{1-x}(ND_4)_xD_2PO_4$. The system is described by a classical pseudospin Ising model with infinite-range exchange interactions and quenched random electric fields. It is shown that in the fast-motion limit the NMR line shape is directly related to the average probability distribution of local deuteron polarization and that the observable truncated second moment of the NMR line M_2^{tr} is proportional to the Edwards-Anderson order parameter q_{EA}. Leading dynamic corrections due to the slowing down of deuteron jumps in the ergodic phase are evaluated, and the behavior of M_2^{tr} at the crossover between the fast- and the slow-motion regimes is discussed.

I. INTRODUCTION

In contrast to magnetic spin glasses, the random freezeout of electric dipole moments in so-called proton or deuteron glasses is characterized by the simultaneous presence of both random exchange interactions and random local electric fields.[1-4] Typical representative systems are solid solutions of hydrogen-bonded ferroelectric and antiferroelectric crystals such as $Rb_{1-x}(NH_4)_xH_2PO_4$ (RADP) and its deuterated counterpart $Rb_{1-x}(ND_4)_xD_2PO_4$ (DRADP).[1] Since the random-field variance Δ acts as an effective ordering field for the glassy state, the corresponding Edwards-Anderson[5,6] order parameter q_{EA} is expected to be nonzero at all temperatures. A static replica theory based on the pseudospin random-bond Ising model with infinite-range interactions,[7,8] where the pseudospin symbolizes the two possible equilibrium sites for a proton (deuteron) in the O—H $\cdots$ O hydrogen bond, predicts that a replica-symmetric proton (deuteron) pseudospin-glass phase is stable in a range of temperatures T and random-field strengths Δ above the instability line $T_I(\Delta)$.[8,9] Alternatively, the instability line can be interpreted within the framework of the dynamic theory of spin glasses[10,11] as a line separating the ergodic from the nonergodic pseudospin glass phase.

It has recently been demonstrated[1] that quadrupole-perturbed nuclear magnetic resonance (NMR) provides a powerful technique to determine the average local-polarization distribution and its second moment, the Edwards-Anderson order parameter in proton and deuteron glasses.[12] In the fast-motion regime at high temperatures, i.e., when the deuteron intrabond jump rate is large compared to the quadrupole splitting, the frequency of the O—D $\cdots$ O deuteron NMR line at a given site is proportional to the local deuteron polarization. Consequently, the second moment of the site-averaged distribution of NMR frequencies is a direct measure of the Edwards-Anderson order parameter q_{EA}. An analogous relationship can be established for the ^{87}Rb NMR line in DRADP (Ref. 1) and the Tl^{2+} EPR spectra in doped RADP.[13] At low temperatures, however, a transition into the slow-motion regime gradually occurs, and, in order to be able to extract quantitative information on the deuteron-glass order parameter from the measured NMR line shape, it is necessary to take into account dynamic effects due to the slowing down of deuteron jumps.

In this paper we present a dynamic theory of the deuteron NMR line shape in deuteron glasses based on the infinitely ranged random-bond Ising model with the addition of quenched local random fields. As argued earlier, this model is—in spite of its simplicity—capable of describing the essential features of the pseudo-spin-glass phase in DRADP systems.[8] We limit ourselves to deuterated systems where the tunneling splitting of the ground-state doublet in the O—D $\cdots$ O potential represents only a small perturbation on an energy scale relevant to the observable dynamic effects. Thus, the transverse field usually associated with the tunneling motion of protons can be neglected, and the system is accordingly described by the classical Ising pseudospin model. To deal with the dynamics of deuteron intrabond jumps, we will apply the method of Glauber kinetics for discrete Ising spins[14] using the recently developed path-integral formulation of Sommers.[15,16] In this approach one can avoid the difficulties associated with the familiar replica formalism while preserving the discrete Ising nature of the dynamic variables at each step in the perturbation expansion.[15] In principle, analogous results can be obtained if one starts from the Langevin equations of motion for continuous soft-spin variables, subsequently transforming back to the correct Ising limit.[10,11]

In Sec. II of this paper we outline the path-integral formalism and its application to the pseudospin model of deuteron glasses (DG). The reader who is unfamiliar with this formalism may skip most of this section on first

reading. In Sec. III, the line shape of quadrupole-unperturbed deuteron NMR is calculated, and in Sec. IV it is related to the local-polarization distribution. Section V deals with the observable second moment of the NMR line which is related to the DG order parameter in the fast-motion limit, and its behavior at the crossover between the fast- and the slow-motion regime is discussed. The results are summarized in Sec. VI. In Appendix A, the local-polarization distribution is calculated using the replica formalism, while in Appendix B the derivation of the truncated second moment of the NMR line is presented.

II. PATH-INTEGRAL FORMALISM FOR DEUTERON GLASSES

The microscopic state of a proton glass (PG) or deuteron-glass system is specified by the vector $\sigma=(\sigma_1,\sigma_2,\ldots,\sigma_N)$, where the pseudospin $\sigma_i=\pm1$ refers to the left-right position of a proton (deuteron) in the ith O—D $\cdots$ O hydrogen bond.[17] By assumption, the time evolution of this state is governed by a Markov process involving random pseudospin flips due to the interaction with a heat bath. (See Fig. 1.) This approach is by now standard[18] and we will only describe the steps necessary to stress the specific features of the present physical problem. First, we introduce the probability $P(\sigma,t)$ for finding the system in a state σ at time t, which obeys the master equation

$$\frac{\partial}{\partial t}P(\sigma,t)=-\sum_{\sigma'}[W(\sigma|\sigma')P(\sigma,t) - W(\sigma'|\sigma)P(\sigma',t)] , \quad (1)$$

where $W(\sigma|\sigma')$ is the rate of transition from state σ to state σ'. In the Glauber model, one focuses on the one-pseudo-spin-flip processes $\sigma_i\rightarrow-\sigma_i$ and the corresponding transition rate is written in a form explicitly satisfying the condition of detailed balance:[15]

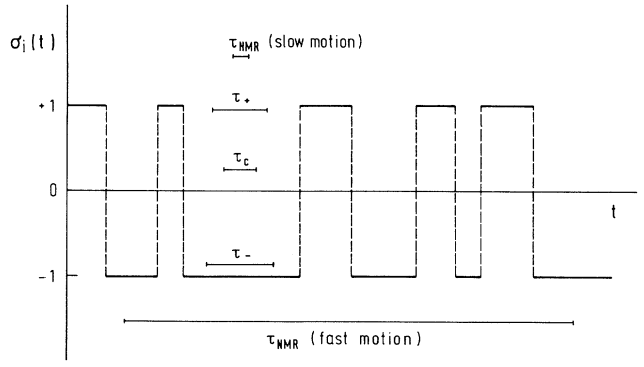

FIG. 1. Schematic time dependence of the local pseudospin variable $\sigma_i(t)$ describing stochastic deuteron motion between two equilibrium sites in the O—D $\cdots$ O bond. Also shown are the average dwell times $\tau_\pm$ and correlation time $\tau_c=1/\Gamma$ defined in Eq. (36). The relation between τ_c and the characteristic time scale $\tau_{\rm NMR}$ for quadrupole perturbed NMR determines the fast- ($\tau_c\ll\tau_{\rm NMR}$) and slow-motion ($\tau_c\gg\tau_{\rm NMR}$) regimes, where $\tau_{\rm NMR}=1/2\omega_1$.

$$W(\sigma_1\cdots\sigma_i\cdots\sigma_N|\sigma_1\cdots-\sigma_i\cdots\sigma_N) = \tfrac{1}{2}\Gamma(1-\sigma_i\tanh\beta h_i) . \quad (2)$$

Here Γ is the pseudo-spin-flip rate and $h_i=h_i(t)$ represents the local field acting on the ith pseudospin, which for a DG system is given by[8]

$$h_i=E_i+f_i+\sum_j J_{ij}\sigma_j \quad (3)$$

with E_i representing the value of an external electric field E at site i, f_i the internal random electric field due to substitutional disorder, and J_{ij} the random interbond coupling. The probability distributions of random bonds and fields are given by two independent Gaussians:

$$P(J_{ij})=\left[\frac{N}{2\pi J^2}\right]^{1/2}\exp(-NJ_{ij}^2/2J^2) ,$$
$$P(f_i)=\frac{1}{\sqrt{2\pi\Delta}}\exp(-f_i^2/2\Delta) . \quad (4)$$

As usual, in the infinite-range model, the scaling of $J_{ij}\propto1/\sqrt{N}$ ensures that the free energy behaves as an extensive variable. In general, the pseudospin-flip rate Γ in Eq. (2) may be depend on the local field h_i and the temperature.

Following Sommers[15] we now rewrite Eq. (1) in the compact form

$$\frac{\partial}{\partial t}P(\sigma,t)=RP(\sigma,t) , \quad (5)$$

where R is the relaxation operator

$$R=\sum_i\tfrac{1}{2}\Gamma(Y_i-1)(1-\sigma_i\tanh\beta h_i) , \quad (6)$$

with Y_i representing the pseudo-spin-reversal operator, i.e.,

$$Y_iF(\sigma_i)=F(-\sigma_i)Y_i$$

for any function F. The formal solution of Eq. (5) is

$$P(\sigma,t)=\hat{T}\exp\left[\int_{t_0}^{t}d\tau R(\tau)\right]P(\sigma,0) , \quad (7)$$

where $\hat{T}$ is the time-ordering operator. It is convenient to choose the initial condition for Eq. (5) in a form corresponding to a system of independent pseudospins[15]

$$P(\sigma,0)=\prod_{j=1}^{N}\tfrac{1}{2}(1+\sigma_jp_j^0) , \quad (8)$$

where p_j^0 is the polarization of bond j at $t=t_0=0$. It may also be noted that, for a system in thermodynamic equilibrium, the stationary solution of Eq. (3) should be canonical, i.e.,

$$P(\sigma)_{\rm eq}=e^{-\beta H}/{\rm Tr}(e^{-\beta H}) \quad (9)$$

with H representing here the model Hamiltonian for a DG system[8]

$$H=-\tfrac{1}{2}\sum_{i,j}J_{ij}\sigma_i\sigma_j-\sum_i(E_i+f_i)\sigma_i . \quad (10)$$

The quantities of interest will be the pseudo-spin-correlation functions

$$\langle \sigma_i(t_1)\sigma_j(t_2)\cdots\sigma_n(t_r)\rangle = \sum_{\sigma} \hat{T} \exp\left[\int_0^t d\tau R(\tau)\right] \sigma_i(t_1)\sigma_j(t_2)\cdots\sigma_n(t_r) P(\sigma,0) \ . \tag{11}$$

This definition includes the local case $i=j=\ldots$, but $t_1 \neq t_2 \neq \ldots$, etc. In general, the correlation functions (11) still depend on the initial polarizations p_j^0.

As first shown by Sommers,[15] the time-ordered product in Eq. (11) can be explicitly evaluated as a path integral over real fields $h_j(\tau)$, $\hat{h}_j(\tau)$, $\sigma_j(\tau)$, and $\hat{\sigma}_j(\tau)$, leading to the result[15,16]

$$\begin{aligned}\langle \sigma_i(t_1)\cdots\sigma_n(t_r)\rangle = \int \prod_{j=1}^{N} Dh_j(\tau) D\hat{h}_j(\tau) D\sigma_j(\tau) D\hat{\sigma}_j(\tau) \sigma_i(t_1)\cdots\sigma_n(t_r) \\ \times \exp\left[-i\int_0^t d\tau \sum_{i,j}^{N} \hat{h}_j(\tau)\left[h_j(\tau) - \sum_i J_{ij}\sigma_i(\tau) - f_j(\tau) - E_j(\tau)\right]\right] \\ \times \exp\left[-i\int_0^t d\tau \sum_{j=1}^{N} \hat{\sigma}_j(\tau)[\sigma_j(\tau) - p_j(\tau)]\right] \ .\end{aligned} \tag{12}$$

Here the fields $p_j(\tau)$ represent local polarizations which obey the exact stochastic equations

$$\frac{\partial p_j}{\partial \tau} = i\hat{\sigma}_j(1-p_j^2) - \Gamma(p_j - \tanh\beta h_j) \tag{13}$$

with initial conditions $p_j(0)=p_j^0$.

Since J_{ij} and f_j appear linearly in the exponent of Eq. (12), the right-hand side (rhs) can be explicitly averaged over both of the random distributions (4). The lowest nontrivial average of the type (12) is related to the bulk pseudospin polarization

$$\bar{p}(t_1) = \frac{1}{N}\sum_i [\langle \sigma_i(t_1)\rangle]_{\rm av} = [\langle \sigma(t_1)\rangle]_{\rm av} \ , \tag{14}$$

where $[\cdots]_{\rm av}$ denotes the simultaneous average over random bonds and random fields. Similarly, the local two-point correlation and response functions are given by

$$\begin{aligned} C(t_1,t_2) &= \frac{1}{N}\sum_i [\langle \sigma_i(t_1)\sigma_i(t_2)\rangle]_{\rm av} \\ &= [\langle \sigma(t_1)\sigma(t_2)\rangle]_{\rm av} \ , \end{aligned} \tag{15a}$$

$$\begin{aligned} G(t_1,t_2) &= \frac{1}{N}\sum_i \frac{\delta[\langle \sigma_i(t_1)\rangle]_{\rm av}}{\delta E_i(t_2)} \\ &= \frac{\delta[\langle \sigma(t_1)\rangle]_{\rm av}}{\delta E(t_2)} \ . \end{aligned} \tag{15b}$$

Equations (14)–(15) imply the self-averaging feature of the DG system in the limit $N\to\infty$.

When the system is in equilibrium, $\bar{p}(t_1)$ in Eq. (14) must be time independent, whereas the functions (15) are expected to depend only on the difference $t=t_1-t_2$. The long-time limit of C then corresponds to the Edwards-Anderson order parameter, i.e., $\lim C(t\to\infty)=q_{\rm EA}$. In the presence of random fields, this quantity is nonzero at all temperatures. In particular, in the ergodic DG phase above the Almeida-Thouless (AT) instability line,[8,9] $q_{\rm EA}$ is equal to the DG order parameter q. Thus, for $T>T_I$, where $T_I=T_I(\Delta)$ is the instability temperature, it is convenient to write[15]

$$C(t) = \tilde{C}(t) + q \ , \tag{16}$$

where $\lim\tilde{C}(t\to\infty)=0$. Likewise, one expects $\tilde{G}(t\to\infty)=0$.

The calculation of the correlation functions (12) in the ergodic phase now parallels that for the magnetic case[15,16] with only one extra contribution to the quadratic terms in the exponent resulting from the averaging over f_j, i.e., $q\to q+\Delta/J^2$, similar to the static replica theory. After linearizing this term by means of a Guassian transformation, one obtains a modified generating functional for the correlation and response functions:

$$\begin{aligned} Z(i\hat{\sigma},E) = \int_{-\infty}^{+\infty} dx \frac{e^{-x^2/2}}{\sqrt{2\pi}} \exp\left[\frac{1}{2}J^2\int\int d\tau_1 d\tau_2 \left[\tilde{C}(1,2)\frac{\delta}{\delta h(1)}\frac{\delta}{\delta h(2)} + 2\Theta(1\text{-}2)\tilde{G}(1,2)\frac{\delta}{\delta h(1)}\frac{\delta}{\delta i\hat{\sigma}(2)}\right]\right] \\ \times \exp\left[\int i\hat{\sigma}(\tau)p(\tau)d\tau\right]\Big|_{h=h(x)} \ , \end{aligned} \tag{17}$$

where Θ denotes the step function,

$$h(x) = E + J(q+\tilde{\Delta})^{1/2}x$$

with $\tilde{\Delta}\equiv\Delta/J^2$, and $p(\tau)$ is the solution of the stochastic equation

$$\begin{aligned}\frac{\partial p(\tau)}{\partial \tau} = i\hat{\sigma}(\tau)[1-p(\tau)^2] \\ -\Gamma[p(\tau) - \tanh\beta h(\tau)] \ .\end{aligned} \tag{18}$$

From Eq. (16) one can obtain any correlation function

$$[\langle \sigma(t_1)\cdots\sigma(t_n)\rangle]_{\rm av}$$

via the corresponding functional derivatives

$$\delta^n Z/\delta i\hat{\sigma}(t_1)\cdots\delta i\hat{\sigma}(t_n)$$

evaluated at $\hat{\sigma}=0$. The response functions can similarly be derived by performing the derivatives with respect to the external field $E(t)$. The unknown two-point functions $\tilde{C}$ and $\tilde{G}$ in Eq. (17) can be determined self-consistently by means of a perturbation expansion. Furthermore, they are related by the fluctuation-dissipation theorem[15]

$$\tilde{G}(t)=-\beta\Theta(t)\frac{\partial\tilde{C}(t)}{\partial t}\ . \tag{19}$$

It can be shown that the static behavior in the ergodic phase is independent of the short-time functions $\tilde{C}(t)$ and $\tilde{G}(t)$. Thus, the correlation and response functions in equilibrium can readily be calculated from Eqs. (17)–(19), and the results agree with the replica theory of DG.

III. LINE SHAPE OF QUADRUPOLE PERTURBED NMR

Since deuterons have a nuclear quadrupole moment, the Larmor frequency ω_L of the ith O—D $\cdots$ O deuteron in an external magnetic field is perturbed by the interaction with the corresponding local electric-field gradient (EFG) tensor. For symmetry reasons, only the orientation—but not the magnitude—of the EFG tensor depends on the position of the deuteron in the O—D $\cdots$ O bond, i.e., for a certain orientation of the bond in the external magnetic field the resonance frequency ω_i is proportional to the instantaneous position $\sigma_i(t)$ of the deuteron

$$\omega_i(t)=\omega_L+\omega_Q+\omega_1\sigma_i(t)=\omega_0+\omega_1\sigma_i(t)\ , \tag{20}$$

where ω_Q and ω_1 are the position-independent and position-dependent contributions of the quadrupole coupling, respectively, and $\omega_0=\omega_L+\omega_Q$. Writing $\omega=\omega_{\rm sig}-\omega_0$, where $\omega_{\rm sig}$ is the frequency of the NMR signal in the laboratory frame, one can characterize the NMR spectrum by the line-shape function[18,19]

$$\begin{aligned} I(\omega)&=\frac{1}{\pi}{\rm Re}\int_0^{+\infty}dt\,e^{-i\omega t}\frac{1}{N}\sum_i\left[\left\langle\exp\left[i\omega_1\int_0^t\sigma_i(\tau)d\tau\right]\right\rangle\right]_{\rm av}\\ &=\frac{1}{\pi}{\rm Re}\int_0^{+\infty}dt\,e^{-i\omega t}\left[\left\langle\exp\left[i\omega_1\int_0^t\sigma(\tau)d\tau\right]\right\rangle\right]_{\rm av}\ . \end{aligned} \tag{21}$$

With the help of the generating functional $Z(i\hat{\sigma},E)$ introduced in Eq. (17), this can be rewritten as

$$\begin{aligned} I(\omega)&=\frac{1}{\pi}{\rm Re}\int_0^{+\infty}dt\,e^{-i\omega t}\exp\left[i\omega_1\int_0^t\frac{\delta}{\delta i\hat{\sigma}(\tau)}d\tau\right]Z(i\hat{\sigma},0)\Big|_{\hat{\sigma}=0}\\ &=\frac{1}{\pi}{\rm Re}\int_0^{+\infty}dt\,e^{-i\omega t}Z(i\omega_1,0)\ , \end{aligned} \tag{22}$$

where the last line follows from the fact that the exponential operator merely translates all variables $i\hat{\sigma}(\tau)$ in Z by the same amount $i\omega_1$. It should be noted that the above relation between the line-shape function $I(\omega)$ and the Glauber-Sommers generating functional Z is still general.

We will now evaluate $I(\omega)$ by neglecting the short-time pseudospin correlations contained in the functions $\tilde{C}$ and $\tilde{G}$. Physically, this is equivalent to assuming that the time scale of the NMR experiment is such that the deuterons can make a very large number of jumps and the system remains close to the equilibrium. Thus, we find from Eqs. (17) and (22)

$$I(\omega)=\frac{1}{\pi}{\rm Re}\int_0^{+\infty}dt\,e^{-i\omega t}\left\langle\exp\left[i\omega_1\int_0^t p_1(\tau)d\tau\right]\right\rangle_x\ , \tag{23}$$

where $\langle\cdots\rangle_x$ denotes a Gaussian average as in Eq. (17), and $p_1(\tau)$ is the solution of a modified Eq. (18)

$$\partial p_1/\partial\tau=i\omega_1(1-p_1^2)-\Gamma[p_1-\tanh\beta h(x)]\ , \tag{24}$$

where $h(x)=J(q+\tilde{\Delta})^{1/2}x$. We will choose the initial condition

$$p_1(0)=p\equiv\tanh\beta h(x)\ ,$$

i.e., at $t=0$ the polarization is given by its equilibrium value in the local field $h(x)$.

The long-time behavior of $p_1(\tau)$ is determined by the complex fixed points of Eq. (24), i.e.,

$$p_\pm(p)=i[\Gamma/2\omega_1\pm r(p)] \tag{25}$$

with $r(p)=[(\Gamma/2\omega_1)^2-1+i\Gamma p/\omega_1]^{1/2}$. The solution of Eq. (24) can then be written in the form

$$p_1(\tau)=(p_- -Bp_+e^{-2\omega_1 r\tau})/(1-Be^{-2\omega_1 r\tau})\ , \tag{26}$$

where $r=r(p)$, $p_\pm=p_\pm(p)$, and the integration constant is given by

$$B\equiv B(p)=(p-p_-)/(p-p_+)\ .$$

We can now easily perform the integration in Eq. (23) and obtain the result for the line-shape function $I(\omega)$:

$$I(\omega)=\frac{1}{\pi}{\rm Re}\int_0^{+\infty}dt\,e^{-i\omega t}\langle A(t)\rangle_x\ , \tag{27}$$

where

$$A(t)=i(\omega_1/r)[(p-p_+)e^{i\omega_1 p_- t}-(p-p_-)e^{i\omega_1 p_+ t}] . \tag{28}$$

IV. LOCAL-POLARIZATION DISTRIBUTION

As argued below, we can identify the quantity $p=\tanh\beta h(x)$ in Eq. (24) as the local pseudospin polarization in a Gaussian random field x. From Eqs. (14) and (17) the equilibrium bulk polarization is given by

$$\bar{p}=[\langle\sigma\rangle]_{\rm av}=\frac{\delta Z}{\delta i\hat{\sigma}(t)}=\int_{-\infty}^{+\infty}dx\frac{e^{-x^2/2}}{\sqrt{2\pi}}\tanh[\beta J(q+\tilde{\Delta})^{1/2}x] , \tag{29}$$

where the derivative has been evaluated for $t\to\infty$ and $\hat{\sigma}=E=0$. The last expression agrees with the result of the static replica theory of proton glasses.[8] It may be noted that $\bar{p}=0$ for a symmetric bond distribution and zero external field E.

By introducing a new integration variable $p=\tanh\beta h(x)$, we can formally rewrite Eq. (29) as

$$\bar{p}=\int_{-1}^{+1}dp\ W(p)p , \tag{30}$$

where

$$W(p)=\frac{1}{\beta J[2\pi(q+\tilde{\Delta})]^{1/2}}\frac{1}{1-p^2}\times\exp\left[-\frac{1}{2}\frac{\operatorname{arctanh}^2 p}{\beta^2J^2(q+\tilde{\Delta})}\right] . \tag{31}$$

The same substitution can now be made to rewrite the average over x in Eq. (27) as an average over p. Inverting the order of integrations in Eq. (27), we obtain

$$I(\omega)=\int_{-1}^{+1}dp\ W(p)I(\omega,p) , \tag{32}$$

where

$$I(\omega,p)=\frac{1}{\pi}\mathrm{Re}\int_0^{+\infty}dt\,e^{-i\omega t}A(p,t) , \tag{33}$$

and the function $A(p,t)$ is given by Eq. (28). Evaluating the integral in Eq. (33) we find, after some simple algebra,

$$I(\omega,p)=\frac{1}{\pi}\frac{\Gamma\omega_1^2(1-p^2)}{(\omega^2-\omega_1^2)^2+\Gamma^2(\omega-\omega_1 p)^2} . \tag{34}$$

The function $I(\omega,p)$ represents the line shape due to the chemical exchange in an asymmetric two-site potential. It contains the effects of time-dependent pseudospin fluctuations around equilibrium in the lowest nontrivial order. At this order, the short-time part of the pseudospin-correlation function is given by its unperturbed value, i.e.,

$$\tilde{C}(t)\cong(1-q)\exp(-\Gamma|t|) .$$

It is easily verified by contour integration that $I(\omega,p)$ is properly normalized, i.e.,

$$\int_{-\infty}^{+\infty}d\omega\,I(\omega,p)=1 . \tag{35}$$

In principle, Eq. (34) could also be derived from the general theory of the NMR line shape.[19] Instead of a single autocorrelation time $\tau_c=1/\Gamma$ for a particle jumping between two sites in a symmetric bistable potential, however, one has to introduce two parameters $\tau_\pm$ corresponding to the average dwell times in the left or right minimum of an asymmetric O—D $\cdots$ O potential with bias energies $\pm\Delta E$, respectively. For a Markov process, the relation between τ_c and $\tau_\pm$ is given by

$$\Gamma=1/\tau_c=\frac{1}{\tau_+}+\frac{1}{\tau_-} , \tag{36}$$

where $\tau_+=\tau_-\exp(-2\beta\Delta E)$ according to the detailed balance condition. This is illustrated schematically in Fig. 1. Furthermore, the local static pseudospin polarization can be written as

$$p=(\tau_--\tau_+)/(\tau_-+\tau_+)=\tanh(\beta\Delta E) . \tag{37}$$

Clearly, in the present deuteron-glass model one has $\Delta E=h(x)$.

Equations (29) and (30) suggest that the function $W(p)$ represents the static equilibrium probability distribution of local polarization p, which is defined as[1]

$$W(p)=\frac{1}{N}\sum_i\delta(p-\langle\sigma_i\rangle)=[\delta(p-\langle\sigma\rangle)]_{\rm av} . \tag{38}$$

It is shown in Appendix A that the random average in Eq. (38) can be evaluated by means of the replica trick, and the result is precisely the same as given by Eq. (31). Here $W(p)$ has been derived within the framework of Glauber-Sommers dynamics in the long-time limit, which is thus indeed equivalent to a static equilibrium.

The second moment of $W(p)$ is, according to Eq. (38),

$$\int_{-1}^{+1}dp\ W(p)p^2=\frac{1}{N}\sum_i\langle\sigma_i\rangle^2=q_{\rm EA} , \tag{39}$$

which is just the Edwards-Anderson order parameter for

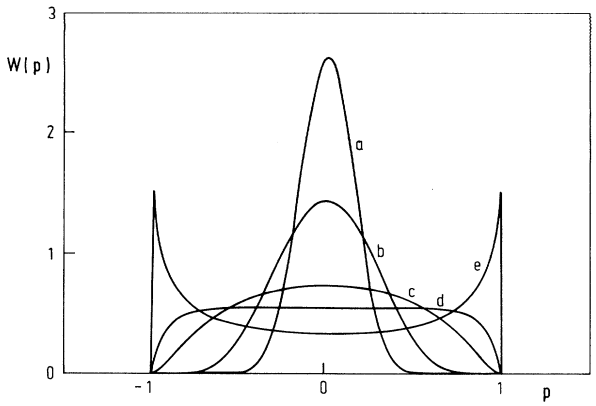

FIG. 2. Local-polarization distribution $W(p)$ calculated for fixed temperature $T=T_G=J/k$ and several values of random-field variance $\tilde{\Delta}$: (a) 10^{-3}, (b) 10^{-2}, (c) 0.1, (d) 0.25, and (e) 1.0.

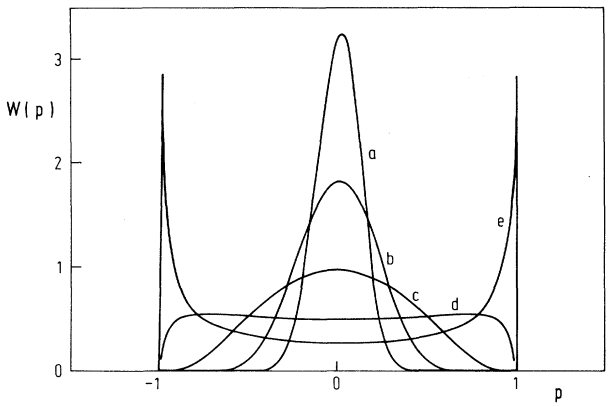

FIG. 3. Same as Fig. 2, but for fixed $\tilde{\Delta}=0.33$ and several values of reduced temperature T/T_G: (a) 9.1, (b) 5.3, (c) 1.0, (d) 0.62, and (e) 0.1.

a DG system. In the ergodic phase one has $q=q_{\rm EA}$. From Eqs. (31) and (39) we recover the self-consistency relation for the DG order parameter[8]

$$q=\int_{-\infty}^{+\infty} dx \frac{e^{-x^2/2}}{\sqrt{2\pi}} \tanh^2[\beta J(q+\tilde{\Delta})^{1/2}x] , \tag{40}$$

which can, of course, also be derived directly from Eq. (17).

In Fig. 2, the local-field probability distribution function $W(p)$ is plotted for $T=T_G$, where T_G is the nominal freezing temperature $T_G=J/k$, and several values of the random-field variance $\tilde{\Delta}$. Similarly, Fig. 3 shows $W(p)$ for $\tilde{\Delta}=0.33$ and several values of the reduced temperature T/T_G. It is evident that, at high temperatures, $W(p)$ is bell shaped with a maximum at $p=0$ which narrows on increasing the temperature. For $T\to\infty$, $W(p)$ approaches a δ function as is readily seen from Eq. (31). For $T<T_G$, however, $W(p)$ exhibits a symmetric two-peak structure with its maxima moving towards $p=\pm 1$ as the temperature is lowered. It is interesting to note that, in the absence of random fields, i.e., for $\Delta\to 0$, one again has $W(p)\to\delta(p)$ for all temperatures $T>T_G$, as it follows from Eq. (31) for $q\to 0$.

V. SECOND MOMENT OF THE NMR LINE

It has been argued in Ref. 1 that Eqs. (32) and (34) can be used to interpret the measured deuteron NMR line shape in DRADP and determine the relevant parameters of the model. Let us therefore examine more closely the relationship between the predicted line-shape function $I(\omega)$ and the observable quantities.

First, we notice from Eqs. (25)–(28) that the character of the line-shape function will generally depend on the ratio of $\Gamma/2\omega_1$, since the function $r(0)$ in Eq. (25) has a branch point at $\Gamma/2\omega_1=1$. Physically, the case $\Gamma>2\omega_1$ corresponds to the so-called *fast-motion regime* where the characteristic NMR time $\tau_{\rm NMR}=1/2\omega_1$ is longer than the correlation time τ_c. By contrast, the case $\Gamma<2\omega_1$ will be referred to as the *slow-motion regime* (cf. Fig. 1).

Next, we want to specify the physical process responsible for the pseudospin-flip rate Γ. A standard model applicable to the case of deuterons in DRADP (Ref. 1) is that of thermally activated jumps across a potential barrier E_a with an additional biasing $\pm h(x)$ due to a coupling to the DG order parameter. Thus, from Eqs. (36) and (37), one obtains

$$\Gamma=\Gamma_0/(1-p^2)^{1/2} , \tag{41}$$

where we assume that Γ_0 obeys the Arrhenius law

$$1/\Gamma_0=\tau_0=\tau_\infty\exp(\beta E_a) , \tag{42}$$

with τ_∞ playing the role of an inverse attempt frequency. Since Γ diverges for $p\to\pm 1$, some of the bonds will always be in the fast-motion regime, even if $\Gamma_0<2\omega_1$. Thus, in general, the line shape $I(\omega)$ might display features corresponding to an average over these two regimes.

A standard method to analyze the experimental NMR line shape is to consider its second moment M_2. In Ref. 1 the second moment of the ^{87}Rb line in $Rb_{0.56}(ND_4)_{0.44}D_2PO_4$, which is simply related to the deuteron line shape, has been used to determine the Edwards-Anderson order parameter $q_{\rm EA}$ in the fast-motion regime. Here we discuss the relation between M_2 and $q_{\rm EA}$ in more detail including the role played by the dynamic effects. It should be noted that, according to the general theory of NMR,[19] the complete second moment is not affected by molecular motion. However, in any NMR experiment the available frequencies are limited, and thus the observed second moment is always truncated, i.e., evaluated over a finite frequency range $-\omega_c<\omega<\omega_c$, where ω_c is some cutoff frequency. Under specific conditions, the truncated second moment $M_2^{\rm tr}$ can be used to determine the molecular rate processes, or in the present case, the DG order parameter.

As shown in Appendix B, the general expression for the truncated second moment can be written in the form

$$M_2^{\rm tr}=(\omega_1^2/2\pi\Omega)\int_{-1}^{+1} dp\, W(p)\frac{\Omega^2-\xi^2}{\xi^2+\eta^2}\int_{-\lambda}^{+\lambda} d(\omega/\omega_1)\left[\frac{\eta^2(\eta^2+\Omega\xi)+(\Omega-\xi)^2(\eta^2-\Omega\xi)}{(\omega/\omega_1-\eta)^2+(\Omega-\xi)^2}\right.$$

$$\left.+\frac{\eta^2(\eta^2-\Omega\xi)+(\Omega+\xi)^2(\eta^2+\Omega\xi)}{(\omega/\omega_1+\eta)^2+(\Omega+\xi)^2}\right] . \tag{43}$$

Here $\Omega=\Gamma/2\omega_1$, $\lambda=\omega/\omega_c$, and the parameters ξ and η represent the real and imaginary part, respectively, of $r(p)$ introduced in Eq. (25), i.e.,

$$r=\xi+i\eta=(\Omega^2-1+2i\Omega p)^{1/2} , \tag{44}$$

with

$$\xi^2=\tfrac{1}{2}(\Omega^2-1+|r|^2) , \tag{45a}$$

$$\eta^2=\tfrac{1}{2}(1-\Omega^2+|r|^2) , \tag{45b}$$

and with $|r|^2=[(\Omega^2-1)^2+4\Omega^2p^2]^{1/2}$. Clearly, Eqs. (43)–(45) are valid both in the fast- ($\Omega>1$) and the slow-motion regime ($\Omega<1$).

The integrals in Eq. (43) are elementary. At the upper limit, they are typically given by

$$(\Omega\pm\xi)^{-1}\arctan[(\lambda\pm\eta)/(\Omega\pm\xi)] ,$$

whereas, at the lower limit, λ is simply substituted by $-\lambda$. Thus, one can easily check, after some rearranging, that the complete second moment, which is obtained for $\lambda\rightarrow\infty$, is precisely equal to

$$M_2=\int_{-\infty}^{+\infty} d\omega\, I(\omega)\omega^2=\omega_1^2 . \tag{46}$$

It should be noted that a typical frequency range for a NMR experiment is $\lambda\cong 3$ or even less. A simple expression for the truncated second moment can be obtained from Eq. (43) in two limiting cases, namely, the *fast-motion limit* ($\Omega\gg 1$) and *slow-motion limits* ($\Omega\ll 1$). Focusing on the *fast-motion limit* first, we derive from Eqs. (45) a power-series expansion for ξ and η in terms of $1/\Omega$:

$$\xi=\Omega-(1-p^2)/2\Omega+O(p^4/\Omega^3) \tag{47a}$$

$$\eta=p+O(p^3/\Omega^2) . \tag{47b}$$

Thus, we see that $\Omega-\xi\ll|\lambda\pm\eta|\ll\Omega+\xi$, and the second integral in Eq. (43) is negligible compared to the first one. After some straightforward algebra and with the help of Eq. (39), we obtain, to leading order,

$$M_2^{\rm tr}=\omega_1^2\int_{-1}^{+1} dp\; W(p)p^2=\omega_1^2 q_{\rm EA} . \tag{48}$$

We conclude that the observable truncated second moment is directly proportional to $q_{\rm EA}$ in the fast-motion limit. Thus, by measuring the second moment of the NMR line, one can determine the temperature dependence of the DG order parameter.[1]

It can easily be shown from Eq. (34) that, in the extreme fast-motion limit, i.e., for $\Omega=\Gamma/2\omega_1\rightarrow\infty$, $I(\omega,p)$ effectively behaves as a δ function:

$$\lim_{\Omega\rightarrow\infty} I(\omega,p)=\delta(\omega-\omega_1 p) , \tag{49}$$

while the line-shape function (33) in the same limit becomes

$$I(\omega)=(1/\omega_1)W(\omega/\omega_1) . \tag{50}$$

Equation (49) suggests that, in the extreme fast-motion limit, a deuteron effectively sees the time-averaged value of the quadrupolar splitting $\omega_1\sigma(t)$, which, for an ergodic system, is equivalent to $\omega_1\langle\sigma\rangle=\omega_1 p$. Thus, Eq. (20) in this limit simplifies to[1]

$$\omega_1=\omega_0+\omega_1\langle\sigma_1\rangle . \tag{51}$$

The resulting line shape can then be interpreted as a static inhomogeneous broadening of the deuteron NMR line due to a random local polarization p having a probability $W(p)$. The inhomogeneous NMR line shape is characterized by the average frequency distribution function[1]

$$f(\omega)=\frac{1}{N}\sum_i \delta(\omega_{\rm sig}-\omega_i)=[\delta(\omega-\omega_1 p)]_{\rm av} , \tag{52}$$

where, again, we write $\omega=\omega_{\rm sig}-\omega_0$. Comparing Eq. (52) with Eqs. (32) and (49), we realize that, in the extreme fast-motion limit, the line-shape function $I(\omega)$ is equivalent to $f(\omega)$. Furthermore, it is trivial to verify from Eq. (52) that the second moment of $f(\omega)$ is equal to $\omega_1^2 q_{\rm EA}$. It should be stressed that this relation is model independent, i.e., it applies both to the long- and the short-range random-bond Ising models of the spin glasses.

In Fig. 4 we compare $I(\omega)$ with $W(\omega/\omega_1)$ for three values of Γ_0/ω_1. For $\Gamma_0\gg 2\omega_1$, the line shape indeed approaches $W(\omega/\omega_1)$, however, for smaller values of Γ_0/ω_1, dynamic effects become increasingly important, resulting in a two-peak structure of $I(\omega)$.

In similar fashion, one can discuss the *slow-motion limit* $\Omega\ll 1$. Equations (45) now yield

$$\xi=\Omega p+O(\Omega^3p^3),\quad \eta=1-\tfrac{1}{2}\Omega^2(1-p^2)+O(\Omega^4) , \tag{53}$$

and it follows that the integrand in Eq. (43) consists of two peaks centered on $\omega/\omega_1\cong\pm 1$ with respective half-widths $\Omega(1\mp p)$. Therefore, the value of the integral is the same as for $\lambda\rightarrow\infty$, and the truncated second moment reaches its maximum value given by $M_2^{\rm tr}=\omega_1^2=M_2$.

For the pseudo-spin-flip model introduced above, this implies that, at low temperatures, where, according to Eqs. (41) and (42), an increasingly large fraction of bonds belongs to the slow-motion regime, one should observe a deviation of $M_2^{\rm tr}$ from its fast-motion value $\omega_1^2 q_{\rm EA}$, and for $T\rightarrow 0$ one expects $M_2^{\rm tr}\rightarrow\omega_1^2$ for all values of ω_1. This is shown in Fig. 5 where the normalized truncated second moment $M_2^{\rm tr}/\omega_1^2$, calculated from Eq. (43) with $\lambda=3$, is

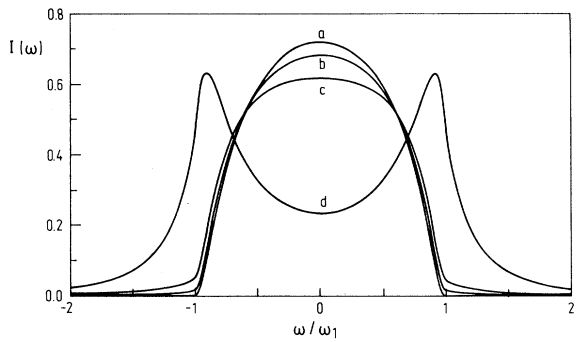

FIG. 4. Line-shape function $I(\omega)$ (in units of ω_1^{-1}) calculated from Eq. (34) for three different values of Γ_0/ω_1, $kT/J=1.33$, and $\tilde{\Delta}=0.5$. (a) $W(\omega/\omega_1)$, (b) $I(\omega)$ for $\Gamma_0/2\omega_1=50$, (c) $\Gamma_0/2\omega_1=1.5$, and (d) $\Gamma_0/2\omega_1=0.05$.

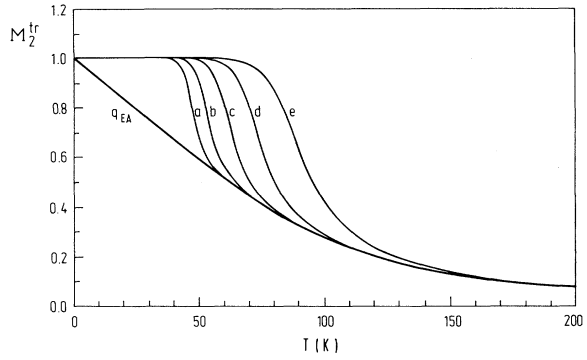

FIG. 5. Calculated temperature dependence of the normalized second moment M_2^{tr}/ω_1^2 for several values of ω_1, illustrating the crossover between the fast- and the slow-motion regimes: (a) $\omega_1/2\pi=10$ kHz, (b) 100 kHz, (c) 1 MHz, (d) 10 MHz, (e) 100 MHz. Heavy line: Edwards-Anderson order parameter q_{EA} calculated from Eq. (40). The model parameters corresponding to DRADP were taken from Ref. 1, i.e., $J/k=90$ K, $\bar{\Delta}=0.35$, and Γ was obtained from Eqs. (41) and (42) with $E_a=80$ meV and $\tau_\infty=1.93\times10^{-14}$ s.

plotted as a function of temperature for five different values of ω_1. The parameter Γ has been obtained from Eqs. (41) and (42) using values of τ_∞ and E_a appropriate to O—D $\cdots$ O deuterons in DRADP, taken from Ref. 1. It is evident that, at high temperatures, the ratio M_2^{tr}/ω_1^2 approaches the fast-motion value q_{EA}; however, in a narrow temperature range, which depends on the value of ω_1, a crossover to the slow-motion limit occurs and, at low temperatures, we find $M_2^{tr}/\omega_1^2\rightarrow1$, as anticipated. The above predictions have been tested by comparing the temperature dependences of M_2^{tr}/ω_1^2 for two probes with different values of ω_1, i.e., for ^{87}Rb where ω_1 is of the order 10^5 Hz, and for Tl^{2+} where $\omega_1\cong10^8$ Hz. A good agreement between the predicted behavior and the experimental data has been found.[1,20]

VI. DISCUSSION AND CONCLUSIONS

In this paper we have presented the calculation of the line shape of quadrupole-perturbed deuteron NMR in structural glasses such as $Rb_{1-x}(ND_4)_xD_2PO_4$. By assumption, the basic line-shape broadening mechanisms are stochastic jumps of deuterons between the two sites in a bistable O—D $\cdots$ O potential. The deuteron jumps are described in terms of a pseudospin Glauber model using the recently developed path-integral formalism by Sommers. In the fast-motion limit at high temperatures, i.e., when the jump rate Γ is much larger than the frequency separation $2\omega_1$ between the resonance frequencies in the left and right equilibrium site in the O—D $\cdots$ O hydrogen-bond potential, the deuteron sees a time-averaged value of the electric-field gradient tensor, which, in the ergodic DG phase, is proportional to the average local pseudospin polarization. Consequently, the NMR line is inhomogeneous broadened by the static probability distribution of local deuteron polarization $W(p)$, and its second moment is a direct measure of the Edwards-Anderson order parameter q_{EA}.

Experiments[1] indicate that, in the low-temperature part of the fast-motion regime, which in DRADP occurs above 45 K, the system is in a crossover regime between the fast- and the slow-motion limits. Therefore, one has to take into account the slowing down of deuteron intrabond jumps, which gives rise to an additional line broadening. These effects have been evaluated here by means of a lowest-order perturbation expansion for the dynamic pseudospin correlation function, leading to a general form for the local chemical exchange line shape $I(\omega,p)$, which depends on the local polarization p as well as the deuteron intrabond jump rate Γ. The resulting NMR line shape can be then obtained by averaging $I(\omega,p)$ over $W(p)$.

By contrast, in the slow-motion limit $\Gamma\ll2\omega_1$, the predicted function $I(\omega,p)$ has the form of a two-peak structure whose maxima are shifted with respect to the unperturbed Larmor frequency ω_0 by the amount $\pm\omega_1$ corresponding to the maximum quadrupolar splitting. In this case, the second moment of the observed NMR line equals its maximum value ω_1^2 independently of q_{EA}. These results demonstrate the importance of taking into account dynamic effects when comparing theoretical and experimental NMR line shapes in glasses.

The theory is easily extended to nuclei other than deuterons such as ^{87}Rb in DRADP. Both deuteron and ^{87}Rb quadrupole-perturbed NMR as well as Tl^{2+} EPR have recently been used to check the validity of some of the predictions, and a good agreement has been found.[1,20] The observed temperature dependence of the Edwards-Anderson order parameter gives strong support to the present model according to which the DG freezing in DRADP is characterized by the presence of both randomly frustrated competing interactions as well as quenched random electric fields created by the substitutional disorder.

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APPENDIX A

Here we derive the probability distribution of local polarization $W(p)$ using the replica formalism. Starting from its definition, Eq. (38), we first rewrite the last average by means of the Fourier representation for the δ function

$$W(p)=\frac{1}{\pi}\mathrm{Re}\int_0^{+\infty}dt\,e^{-ipt}[\exp(i\langle\sigma_i\rangle t)]_{\mathrm{av}}\ . \tag{A1}$$

Next we expand the second exponent into a power series and find

$$W(p)=\frac{1}{\pi}\mathrm{Re}\int_0^{+\infty}dt\,e^{-ipt}\sum_{m=0}^{\infty}i^m t^m[\langle\sigma_i\rangle^m]_{\mathrm{av}}/m!\ . \tag{A2}$$

According to the replica formalism,[6,7] the product of m thermodynamic averages $\langle \hat{O} \rangle$ of any operator $\hat{O}$ can formally be written as a single trace

$$\langle \hat{O} \rangle^m = \lim_{n \to 0} \mathrm{Tr}_n \left[\hat{O}_{\alpha_1} \hat{O}_{\alpha_2} \cdots \hat{O}_{\alpha_m} \exp \left[-\beta \sum_{\alpha=1}^{n} H_\alpha \right] \right] , \tag{A3}$$

where the dummy replica indices $\alpha_1 \cdots \alpha_m$ are all distinct but contained in the sum over the replicated Hamiltonian (10). This is readily applied to the case $\hat{O} = \sigma_1$ in Eq. (A2) and the random average evaluated by integrating over the random distributions (4). One can then perform the standard manipulations from the theory of spin[6,7] and proton[8] glasses with infinite-range interactions. After averaging over the random distributions (4) the result does not depend on the site index i and we obtain in zero external field ($E_i = 0$)

$$[\langle \sigma_i \rangle^m]_{\mathrm{av}} = \lim_{n \to 0} \mathrm{Tr}_n \left[\sigma_{\alpha_1} \sigma_{\alpha_2} \cdots \sigma_{\alpha_m} \exp \left[\frac{1}{2} \beta^2 J^2 \sum_{\alpha \neq \beta} (q_{\alpha\beta} + \tilde{\Delta}) \sigma_\alpha \sigma_\beta \right] \right] , \tag{A4}$$

where $q_{\alpha\beta}$ is the usual spin-glass order parameter. As discussed by Binder and Young,[6] in the case of broken replica symmetry, the rhs of Eq. (A4) has to be averaged over all permutations of the replicas $\alpha_1 \cdots \alpha_m$. In the present problem, however, we focus on the replica-symmetric solution $q_{\alpha\beta} = q$ which is stable above the instability line $T_I(\Delta)$. The bilinear terms $\sigma_\alpha \sigma_\beta$ in the exponent can then be decoupled in the usual manner by introducing a Gaussian random variable x, thus factorizing the trace into a product of n traces over independent replicas. After the limit $n \to 0$ has been performed the average (A4) becomes

$$[\langle \sigma_i \rangle^m]_{\mathrm{av}} = \int_{-\infty}^{+\infty} dx \frac{e^{-x^2/2}}{\sqrt{2\pi}} \tanh^m [\beta J (q + \tilde{\Delta})^{1/2} x] . \tag{A5}$$

Returning to Eq. (A2), we find, after evaluating the sum over m and the integral over t,

$$W(p) = \int_{-\infty}^{+\infty} dx \frac{e^{-x^2/2}}{\sqrt{2\pi}} \delta(p - \tanh[\beta J (q + \tilde{\Delta})^{1/2} x]) . \tag{A6}$$

The remaining integration can be carried out after applying the standard relation

$$\delta(f(x)) = \delta(x - x_0) / |f'(x_0)| ,$$

where $f' = df/dx$ and x_0 is the solution of $f(x) = 0$, leading to the result (31).

APPENDIX B

Here we present a brief derivation of the truncated second moment M_2^{tr} as given by Eq. (43). Introducing a dimensionless parameter $\Omega = \Gamma / 2\omega_1$, we first combine Eqs. (32) and (34) to obtain

$$M_2^{\mathrm{tr}} = 2\Omega\omega_1^2 \int_{-1}^{+1} dp \, W(p)(1 - p^2) K_2 , \tag{B1}$$

where

$$K_2 = \frac{1}{\pi} \int_{-\lambda}^{+\lambda} d(\omega/\omega_1) \frac{(\omega/\omega_1)^2}{[(\omega/\omega_1 - \eta)^2 + (\Omega - \xi)^2][(\omega/\omega_1 + \eta)^2 + (\Omega + \xi)^2]} , \tag{B2}$$

with ξ and η defined in Eqs. (44)–(45). The integrand in Eq. (B2) has four simple poles located at $z_{1,2} = \eta \pm i(\Omega - \xi)$ and $z_{3,4} = -\eta \pm i(\Omega + \xi)$, where $z = \omega/\omega_1$. Thus, it can be decomposed into a sum of partial fractions of the type $\sum_{i=1}^{4} c_i / (z - z_i)$, where c_i is the residue at z_i, e.g.,

$$c_1 = z_1^2 / (z_1 - z_2)(z_1 - z_3)(z_1 - z_4) ,$$

etc. Recombining partial fractions into appropriate pairs, we find

$$K_2 = \frac{1}{4\pi} [(\xi^2 + \eta^2)(\Omega^2 + \eta^2)]^{-1} \int_{-\lambda}^{+\lambda} dz \left[\frac{\eta^2(\eta^2 + \Omega\xi) + (\Omega - \xi)^2(\eta^2 - \Omega\xi)}{(z - \eta)^2 + (\Omega - \xi)^2} + \frac{\eta^2(\eta^2 - \Omega\xi) + (\Omega + \xi)^2(\eta^2 + \Omega\xi)}{(z + \eta)^2 + (\Omega + \xi)^2} \right.$$
$$\left. + \eta(\Omega^2 + \xi^2 + \eta^2) \left[\frac{z - \eta}{(z - \eta)^2 + (\Omega - \xi)^2} + \frac{z + \eta}{(z + \eta)^2 + (\Omega + \xi)^2} \right] \right] . \tag{B3}$$

The last term in large parentheses is an odd function of z and its integral vanishes. After inserting the remaining part of (B3) into Eq. (B1), using the relation

$$(\Omega^2 - \xi^2)(\Omega^2 + \eta^2) = \Omega^2 (1 - p^2) ,$$

and replacing z again by ω/ω_1, we arrive at Eq. (43).

[1]R. Blinc, J. Dolinšek, R. Pirc, B. Tadić, B. Zalar, R. Kind, and O. Liechti, Phys. Rev. Lett. **63**, 2248 (1989).
[2]R. Blinc, D. C. Ailion, B. Günther, and S. Žumer, Phys. Rev. B **30**, 85 (1984).
[3]R. Kind, O. Liechti, and M. Mohr, Ferroelectrics **78**, 87 (1988).
[4]R. A. Cowley, T. W. Ryan, and E. Courtens, Z. Phys. B **65**, 181 (1986).
[5]S. F. Edwards and P. W. Anderson, J. Phys. F **5**, 965 (1975).
[6]For a review see, e.g., K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986).
[7]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. **35**, 1792 (1975).
[8]R. Pirc, B. Tadić, and R. Blinc, Phys. Rev. B **36**, 8607 (1987).
[9]J. R. L. de Almeida and D. J. Thouless, J. Phys. A **11**, 983 (1978).
[10]H. Sompolinsky and A. Zippelius, Phys. Rev. B **25**, 6860 (1982).
[11]B. Tadić, R. Pirc, and R. Blinc, Z. Phys. B **75**, 249 (1989).
[12]A similar relationship has been found earlier in the NMR spectra of H_2 orientational glass by D. Esteve, M. Devoret, and N. S. Sullivan, J. Phys. C **15**, 5455 (1982).
[13]P. Cevc, B. Zalar, and R. Blinc, Solid State Commun. **70**, 461 (1989).
[14]R. J. Glauber, J. Math. Phys. (N.Y.) **4**, 294 (1963).
[15]H.-J. Sommers, Phys. Rev. Lett. **58**, 1268 (1987); (private communication).
[16]For a recent application to the problem of neural networks see H. Rieger, M. Schreckenberg, and J. Zittartz, Z. Phys. B **72**, 523 (1988); **74**, 527 (1989).
[17]See, e.g., R. Blinc and B. Žekš, *Soft Modes in Ferroelectrics and Antiferroelectrics* (North-Holland, Amsterdam, 1974), and references therein.
[18]S. Dattagupta, *Relaxation Phenomena in Condensed Matter Physics* (Academic, London, 1987).
[19]A. Abragam, *The Principles of Magnetic Resonance* (Oxford, New York, 1960), Chap X, p. 447ff.
[20]R. Kind, R. Blinc, J. Dolinšek, N. Korner, B. Zalar, P. Cevc, N. S. Dalal, and J. DeLooze, following paper, Phys. Rev. B **43**, 2511 (1991).