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Coexistence of the critical slowing down and glassy freezing in relaxor ferroelectrics

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We have developed a dynamical model for the dielectric response in relaxor ferroelectrics that explicitly takes into account the coexistence of the critical slowing down and glassy freezing. The application of the model to the experiment in $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) allowed for the reconstruction of the nonequilibrium spin-glass state order parameter and its comparison with the results of recent NMR experiment [R. Blinc *et al.*, Phys. Rev. Lett **83**, 424 (1999)]. It is shown that the degree of the local freezing is rather small even at temperatures where the field-cooled permittivity exceeds the frequency-dependent permittivity by an order of magnitude. This observation indicates the significant role of the critical slowing down (accompanying the glass freezing) in the system dynamics. Also, the theory predicts an important interrelationship between the frequency-dependent permittivity and the zero-field-cooled permittivity, which proved to be consistent with the experiment in PMN [A. Levstik *et al.*, Phys. Rev. B **57**, 11 204 (1998)].

I. INTRODUCTION

Relaxor ferroelectrics, which are disordered perovskite ferroelectrics like $PbMg_{1/3}Nb_{2/3}0_3$ (PMN) or $PbSc_{1/2}Ta_{1/2}O_3$, represent a new class of materials that have been a subject of numerous investigations (see, for example, Refs. 1–13). Relaxor ferroelectrics manifest themselves in the extraordinary low-frequency dispersion of their dielectric permittivity compared with regular ferroelectrics. The position and the height of the permittivity maximum plotted as a function of temperature depends on the frequency of the probe field and shifts to lower temperatures when the frequency decreases.

The latter behavior is accompanied by the observed splitting between the field-cooled (FC) and zero-field-cooled (ZFC) permittivity and the existence of long-lived remanent polarization. However, as has been emphasized recently, the bulk of the relaxation spectra in relaxors remains active even far below the temperature T_f where the FC and ZFC permittivity split, and the nonlinear susceptibility does not diverge at T_f. All these findings indicate nonequilibrium phenomena and quasinonergodicity (''freezing''), rather than a true thermodynamic dipole spin-glass transition.

On the other hand, the existence of the very high dielectric constant indicates that these systems are close to ferroelectric instability, and, therefore, one could expect the manifestation of the critical slowing down of dynamics and a competition between the critical slowing down and the dipole spin-glass freezing. Such a dichotomy makes it a nontrivial task to extract from the experiment quantitative characteristics of the freezing.

One of the important characteristics of spin-glass freezing is a value of the spin-glass order parameter q, which in relaxors is a time-dependent quantity. We will show that this quantity can be extracted from the experimental results on the frequency-dependent permittivity, with the use of the model discussed below. Also the theory predicts an important interrelationship between the frequency-dependent permittivity and the zero-field-cooled permittivity, which proved to be consistent with the experiment.

II. MODEL

It has been proposed recently[12] that relaxor behavior is a common characteristic of the collective dynamics of localized giant dipole moments distributed in highly polarizable lattices. This assumption is consistent with the observation in relaxors of the quasisoft optical modes (remaining finite at all temperatures), frequency of which ω_0 is of the same order of magnitude ($\omega_0 \sim 50\ \mathrm{cm}^{-1}$) as those in highly polarizable dielectrics such as $KTaO_3$ and $SrTiO_3$. The necessary condition of relaxor behavior is the simultaneous existence of the broad distribution of the local field and the broad distribution of dipole relaxation frequencies. Also, experiments[10,11,4] favor the hypothesis that the physical origin of localized dipole moments is the off-center shift of the atom even at high temperatures. In this model, the large values of the localized dipole moment d^* induced by an off-center ion are associated with the polarization cloud (cluster) formed by the simultaneous displacements of the other atoms adjacent to a given off-center ion. The value of d^* determines the energy V_{cl} of the isolated cluster in the applied electric field E_{ex}; $V_{cl} = -d^* E_{ex}$. The basic physical principles governing the formation of localized (giant) dipole moments in highly polarizable crystals (such as doped $KTaO_3$ or $SrTiO_3$) have been reviewed in Ref. 14. It has been proven, also, that the anomalous collective behavior of such crystals is caused by the modification of the dipole-dipole interactions due to the effect of the spatial dispersion of lattice permittivity.

In relaxor ferroelectrics, the notion of polar clusters is more complicated than described above due to strong interactions of closely separated off-center ions. An attempt was made[15] to develop a microscopic theory of cluster formation in highly polarizable materials with the account of pair interactions between off-center ions (dilute limit). This approach has been applied successfully to $K_{1-x}Li_xTaO_3$ ($x < 0.04$).[7] On the other hand, in disordered perovskite relaxors, the concentration of off-center ions is not small and the interactions between them are unknown. In this situation, it seems practical to develop a phenomenological approach of dielectric response that would take into account two main factors distinguishing these systems from conventional ferroelectrics,

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namely, the existence of a broad distribution of local fields and a broad distribution of relaxation times characterizing cluster reorientations.

A convenient approach to describe dynamical behavior of relaxor ferroelectrics is to start from the Bloch-type equations widely explored in the theory of regular ferroelectrics possessing Debye relaxation.[16] We write the Bloch equation in the form

$$\frac{\partial\langle \mathbf{d}^*(t)\rangle}{\partial t}=-\frac{1}{\tau}\,[\langle \mathbf{d}^*(t)\rangle-\langle \mathbf{d}^*\rangle^{eq}_{E(t)}], \tag{1}$$

where $\langle \mathbf{d}^*(t)\rangle$ is the nonequilibrium thermal average value of the effective dipole moment of a polar cluster characterized by the reorientation frequency τ^{-1} and the effective local field $\mathbf{E}(t)$.

Equation (1) describes the relaxation of the dipole moment of each cluster to its quasiequilibrium value $\langle \mathbf{d}^*\rangle^{eq}_{E(t)}$, which depends on the value of the local field $\mathbf{E}(t)$ induced by other clusters at any moment of time[16]

$$\langle \mathbf{d}^*\rangle^{eq}_{E}=\frac{\mathrm{Tr}\,\mathbf{d}^* e^{-\mathbf{d}^*E}/kT}{\mathrm{Tr}\,e^{-\mathbf{d}^*E}/kT}. \tag{2}$$

If, e.g., each dipole moment can be oriented only along two opposite directions, then $\langle d^*\rangle^{eq}_{E}=d^*\tanh(d^*E/kT)$. In Eq. (1) the local field $\mathbf{E}$ is a time-dependent random field. It includes the contribution from other clusters, the contribution from the applied field $\mathbf{E}_{ex}$, and the contribution from the static random field caused by material imperfections. Note that Eq. (1) is quite general and, although the explicit form of $\langle \mathbf{d}^*\rangle^{eq}_{E}$ and the precise definition of the polar clusters are model dependent, it affects only the coefficients of the theory discussed below.

The observable quantity in the dielectric measurements is the average cluster polarization

$$\mathbf{P}(t)=n\overline{\langle \mathbf{d}^*(t)\rangle}\approx n\int d\mathbf{E}\,f(\mathbf{E})\int_{\tau_0}^{\tau_m}d\tau\,g(\tau)\langle \mathbf{d}^*(t)\rangle, \tag{3}$$

where n is the concentration of localized dipole moments and the overbar denotes the configurational average, which includes the average over E and τ. In Eq. (3), $f(\mathbf{E})$ is the distribution function of the local fields, and $g(\tau)$ is the distribution function of relaxation times. As a first step in understanding the complex behavior of relaxor materials, we neglected in Eq. (3) the possible correlation effects between E and τ.

We assume that the distribution function of the local field can be written in the form

$$f(\mathbf{E})\equiv f(\mathbf{E},\mathbf{P})=\tilde{f}(\mathbf{E}-\gamma\mathbf{P}-\mathbf{E}_{ex}). \tag{4}$$

The proposed form of $f(\mathbf{E},\mathbf{P})$ is consistent with that given by the mean-field approximation (δ is the Dirac δ function)

$$f^{MF}(\mathbf{E},\mathbf{P})=\delta(\mathbf{E}-\gamma\mathbf{P}-\mathbf{E}_{ex}), \tag{5}$$

meaning that the local field is equal to $E=\gamma P+E_{ex}$. The effect of the local-field fluctuations corresponds to the replacement of the δ function by the function $\tilde{f}$ (4) of finite width, which depends parametrically on the average polarization P. A self-consistent approach for evaluating $f(\mathbf{E},\mathbf{P})$ in disordered ferroelectrics is discussed in Ref. 14.

The phenomenological parameter γ introduced above characterizes the strength of cluster-cluster interactions. Note that in impurity-induced relaxors such as $\mathrm{K}_{1-x}\mathrm{Li}_x\mathrm{TaO}_3$, where the interaction between localized dipole moments is mediated by soft optical phonons, $\gamma=4\pi/\epsilon_l$ (Ref. 14) (ϵ_l is the background permittivity describing the dielectric response of the highly polarizable lattice). It is apparent that in disordered complex perovskites, such as PMN, PST, etc., the physical picture of cluster-cluster interaction is much more complicated. Therefore, below we will consider γ as a phenomenological fitting parameter. Note also, to avoid confusion, that γ is not a parameter of the Lorentz local-field corrections, since E is a field that experiences an effective dipole moment d^* rather than true microscopic dipole moment $\mathbf{d}$.

It is known that the existence of the broad distribution of relaxation times leads to non-exponential behavior in the polarization relaxation. In order to reproduce this effect within the proposed formalism, we first rewrite Eq. (1) in the integral form

$$\langle d^*(t)\rangle=\langle d^*(0)\rangle e^{-t/\tau}+\frac{1}{\tau}\int_0^t dt'\,e^{-t'/\tau}\langle d^*\rangle^{eq}_{E(t-t')}, \tag{6}$$

and then take the average with respect to τ, and E. Thus, we obtain [note that $(1/\tau)e^{-t/\tau}=-(d/dt)e^{-t/\tau}$]

$$\mathbf{P}(t)=\mathbf{P}(0)q(t)-\int_0^t dt'\frac{dq(t')}{dt'}\mathbf{P}^{eq}(t-t'), \tag{7}$$

where

$$\mathbf{P}^{eq}(t)=n\int d\mathbf{E}\langle \mathbf{d}^*\rangle_E f(\mathbf{E},\mathbf{P}(t)). \tag{8}$$

At $E_{ex}=0$, Eq. (8) determines the values of spontaneous polarization $\mathbf{P}^s$ (Ref. 14)

$$\mathbf{P}^s=n\int d\mathbf{E}\langle \mathbf{d}^*\rangle_E f(\mathbf{E},\mathbf{P}^s). \tag{9}$$

The variable $q(t)$ in Eq. (7) is equal to

$$q(t)=\int_{\tau_0}^{\tau_m}d\tau\,\tilde{g}(\tau)e^{-t/\tau}\approx\int_t^{\tau_m}d\tau\,\tilde{g}(\tau), \tag{10}$$

where the right-hand side expression of Eq. (10) is valid for smooth functions $\tilde{g}(\tau)$. The variable $q(t)$ describes the fraction of clusters effectively frozen at time t and therefore has the meaning of the spin-glass order parameter on a finite time scale. Note that, as we assumed above, $q(t)\rightarrow 0$ at $t\rightarrow\infty$.

Equations (7) and (8) can be applied for the analysis of different experimental situations related to linear and nonlinear polarization responses on the applied field. In this paper, we will concentrate on the analysis of linear dielectric permittivities, assuming that the values of E_{ex} are sufficiently small such that the linear response theory is valid. This assumption is consistent with the body of experiments on linear dielectric permittivity in relaxors. We will consider the temperatures $T>T_c$, where T_c is the temperature of the ferroelectric phase transition.

Expanding the right-hand side of Eq. (7) in the power series with respect to E_{ex} and P and considering the first nonvanishing terms we obtain

$$P(t)=P(0)q(t)-k(T)\int_0^t dt'\frac{dq(t')}{dt'}[\gamma P(t-t')+E_{ex}(t-t')], \tag{11}$$

where the function $k(T)$ is given by

$$k(T)=-n\int dE\langle d^*\rangle_E^{eq}\frac{\partial f(E,P)}{\partial E}\bigg|_{P=0.} \tag{12}$$

III. FREQUENCY-DEPENDENT PERMITTIVITY

We assume that at $t=0$ the system is in thermal equilibrium, meaning that at $T>T_c, P(0)\equiv P_s=0$. The steady-state frequency-dependent permittivity can be easily obtained from Eq. (11) using $E_{ex}(t)=E_{ex}^{(1)}e^{i\omega t}$ and the definition

$$\epsilon(\omega,T)=4\pi\frac{\partial P(\omega)}{\partial E_{ex}^{(1)}}+\epsilon_l. \tag{13}$$

Using the Laplace transform in Eq. (11) we obtain

$$\epsilon(\omega,T)=\frac{4\pi k(T)Q(\omega,T)}{1-\gamma k(T)Q(\omega,T)}+\epsilon_l, \tag{14}$$

where

$$Q(\omega,T)=\int_0^\infty dt\, e^{-i\omega t}\frac{dq}{dt}. \tag{15}$$

For the smooth function $g(\tau)$, the real part of $Q(\omega,T)$ can be simplified as[17]

$$Q'(\omega,T)=\int_{\tau_0}^{\tau_m}d\tau\frac{g(\tau)}{1+\omega^2\tau^2}\approx\int_{\tau_0}^{1/\omega}d\tau\, g(\tau)=1-q(\omega^{-1},T). \tag{16}$$

The relation (16) between the functions $q(t)$ and $Q'(\omega)$ will be employed below to obtained information on the degree of local freezing in relaxors from the frequency-dependent dielectric constant using the fact that usually $Q''<Q'$.

At $\omega=0$, Eq. (14) defines the static or field-cooled permittivity

$$\epsilon_{FC}=\frac{4\pi k(T)}{1-\gamma k(T)}+\epsilon_l. \tag{17}$$

On the other hand, at $\omega\tau_m\gg 1$ the function $Q(\omega,T)\to 0$ and therefore $\epsilon(\omega)\to\epsilon_l$. Thus, in our model ϵ_l can be regarded as a high-frequency permittivity ϵ_∞ (with respect to the dielectric measurements at frequencies $\omega\ll\omega_0$). Combining Eqs. (14) and (17) we obtain

$$Q(\omega)=\frac{\epsilon(\omega)-\epsilon_0}{\epsilon_{FC}-\epsilon_0}\,\frac{1+\frac{\gamma}{4\pi}(\epsilon_{FC}-\epsilon_l)}{1+\frac{\gamma}{4\pi}[\epsilon(\omega)-\epsilon_l]}. \tag{18}$$

Note that in a number of recent publications on relaxors[18,19,13] the analysis of the relaxation spectrum $Q(\omega)$ was based on a different relation between $Q(\omega)$ and the permittivity, namely,

$$Q(\omega)=\frac{\epsilon(\omega)-\epsilon_l}{\epsilon_{FC}-\epsilon_l}, \tag{19}$$

where we substituted ϵ_l for ϵ_∞. Equation (19) was introduced earlier in spin glasses, assuming that the relaxation time τ in Eq. (16) is a characteristic of independent cluster relaxators. However, this equation has a different meaning when applied to relaxor ferroelectrics. Indeed, Eq. (19) can be obtained from Eq. (1) if we first perform the average over the distribution of the local random fields at a constant value of τ and then perform the average over τ. With such a two-step averaging, we arrive at Eqs. (19) and (16) with τ replaced by $\tau^*=\tau\epsilon/\epsilon_l$. Note that τ^* is a relaxation time of the long-wavelength collective polar mode undergoing the critical slowing down of dynamics, rather than the relaxation time of the individual dipoles or clusters. Thus, the applicability of Eqs. (19) and (16) to a system with long-range or mesoscopic polar order implies that the crystal can be divided into macroregions within which the relaxation time of all dipole moments has the same magnitude, and the average over τ means the average of the dielectric response for different macroregions. On the other hand, Eq. (18) is consistent with the formation of the short-range clusters in the absence of correlations between the relaxation times of different clusters.

IV. ZERO-FIELD-COOLED SUSCEPTIBILITY AND REMANENT POLARIZATION

The values of $\epsilon_{ZFC}(t)$ can be obtained by solving Eq. (11) with $P(0)=0$ and $E_{ex}=\text{const}$. We consider here a particular case when one can approximately neglect the memory effects in Eq. (2) by replacing $P(t-t')\approx P(t)$ (which implies a fast decay of dq/dt). Thus, we obtain

$$\epsilon_{ZFC}(t)=\frac{4\pi k[1-q(t)]}{1-\gamma k[1-q(t)]}+\epsilon_l. \tag{20}$$

One can see that $\epsilon_{ZFC}(t)$ is identical to $\epsilon(\omega)$ given by Eq. (14), if one substitutes ω by $1/t$ in Eq. (16).

Equation (11) can be used also for the analysis of the polarization decay at $T>T_c$ [assuming that the initial nonequilibrium polarization $P(0)$ is small enough, such that the linear equation (11) is valid]. In order to obtain the remanent polarization $P_r(t)$, we solve Eq. (11) in the same manner as above, but with $E_{ex}=0$ and $P(0)\neq 0$. We obtain

$$\frac{P(t)}{P(0)}=\frac{q(t)}{1-\gamma k[1-q(t)]}=\frac{\epsilon_{FC}-\epsilon_{ZFC}}{\epsilon_{FC}-\epsilon_l}. \tag{21}$$

The denominator in Eq. (21) reproduces the effect of the critical slowing down of dynamics, which is imposed on the effect of glass freezing characterized by the slow decaying function $q(t)$. For example, when $\gamma k \rightarrow 1$ (i.e., the system is in the vicinity of the second-order phase-transition temperature), it follows from Eq. (21) that $P_r(t) = \text{const}$, independently of the value of $q(t)$.

V. COMPARISON WITH THE EXPERIMENT IN PMN

A. Nonequilibrium spin-glass order parameter

The nonequilibrium spin-glass order parameter $q(t,T)$ is a very important quantity determining the dielectric response of relaxors. The dielectric permittivity can be formulated in terms of the parameter $q(t,T)$ in a way consistent with the description of magnetic susceptibility in magnetic alloys where spin-glass and ferromagnetic order coexist. It is convenient to rewrite Eq. (14) in the identical form with the use of Eq. (16),

$$\epsilon(\omega,T) = \frac{4\pi k(1-q)}{1-\gamma k(1-q)} + \epsilon_l . \tag{22}$$

The first term in Eq. (22) is very similar to the well-known solution for the susceptibility given by the infinite range Sherrington-Kirkpatrick model (mean-field theory)

$$\chi = \frac{C(1-q)}{T-\theta(1-q)} . \tag{23}$$

Equation (23) has been widely used for the description of the experiments on ZFC susceptibility, as well as frequency-dependent susceptibility in magnetic alloys[20] and relaxors,[21] by treating the parameters C and θ as purely phenomenological fitting parameters. In fact, our approach justifies and generalizes Eq. (23) in the case of a nonequilibrium spin-glass state. The deviation of the parameter $k(T)$ from θ/T indicates the deviation from the mean-field picture.

Equations (14) and (16) allow for reconstructing the values of the functions $q(t,T)$ from experiments on the frequency-dependent permittivity. We applied them to the recent data on PMN by Levstik *et al.*,[13] who obtained the values of the frequency-dependent permittivity, as well as field-cooled and zero-field-cooled permittivity. We used $\epsilon_l \approx 100$, corresponding to the values of ϵ at $\omega = 10^{12}$ Hz[13], and $\gamma = 10^{-2}$, provided the good accuracy in reproducing the frequency-dependent permittivity in the temperature range $T = 200$–350 K (see Sec. V B).

The results of the reconstruction are presented in Fig. 1, where the temperature dependences of $q(T)$ are shown for three distinct values of time corresponding to the frequencies of the applied field 20 Hz, 1 kHz, and 100 kHz used in the experiment. One can see that the values of $q(T)$ are rather small even at $T = 200$ K, where the values of $\epsilon(\omega)$ are almost 10 times less than the values of ϵ_{FC}. The explanation of this important effect lies in the extremely high dielectric constant of PMN leading to $\gamma k \approx 0.98$ at $T = 200$ K. Thus, the system is extremely close to ferroelectric instability, which results in the significant effect of the critical slowing down, accompanying the glass freezing, upon the system dynamics.

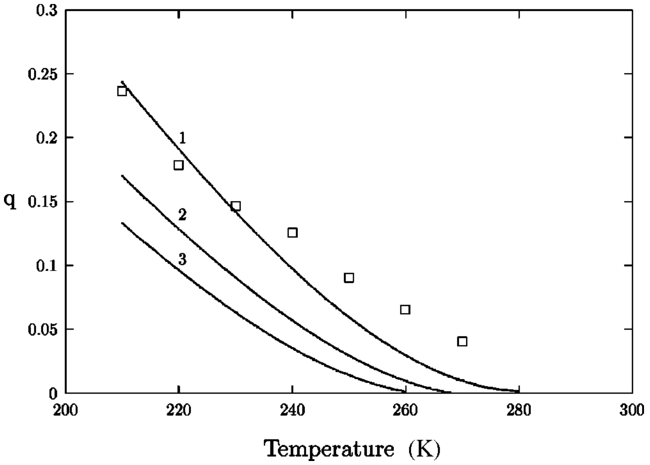

FIG. 1. Reconstructed values of the nonequilibrium spin-glass order parameter in PMN as a function of temperature corresponding to the following frequencies of the applied field: 100 kHz (1), 1 kHz (2), and 20 Hz (3). Squares reproduce the data obtained in Ref. 19.

In Fig. 1, the reconstructed values of q are compared with the data obtained from the recent experiments by Blinc *et al.*[22] on the observation of inhomogeneous broadening of NMR lines of Nb ions. The appearance of inhomogeneous broadening signals the slowing down of the dynamics on the scale of inhomogeneous linewidth (equal to 10–50 kHz). A reasonable correspondence between curve 1 and the data[22] (with the account of comparable time scales in both experiments) allows us to suggest the following scenario for the NMR broadening mechanism in PMN. As known, e.g., from the analysis of nuclear spin-lattice relaxation of Li and Nb ions in KTaO_3:Li,Nb,[14] the appearance of polarization on the mesoscopic length scale in highly polarizable crystals results in a significant modulation of the electric-field gradient, which leads to the modulation of nuclear-resonance frequency. Since in disordered PMN crystals, all atoms lack a center of symmetry, one can assume that the nuclear frequency shift for each Nb atom will be proportional to the corresponding value of the local field $\mathbf{E}$. As a result, the width δ of the inhomogeneous line will be proportional to the fraction of polar clusters [given by the nonequilibrium spin-glass parameter $q(T)$], whose dynamics is effectively frozen on the time scale of $1/\delta$. The above broadening mechanism gives also a natural explanation of the Gaussian line shape observed in Ref. 22, since the distribution function $f(\mathbf{E})$ of the local field is close to Gaussian in highly polarizable crystals,[14] and the shape of the inhomogeneous NMR line coincides with the shape of $f(\mathbf{E})$ for atoms lacking center of symmetry.

B. Interrelationship between ZFC and frequency-dependent permittivity

Another practical aspect of the proposed model, which helps to clarify the experimental situation in PMN, is the interrelationship between the frequency-dependent permittivity and zero-field-cooled permittivity obtained in Sec. IV. In order to employ this interrelation, note first that in PMN, at the temperatures near the temperature of the permittivity maximum, the characteristic relaxation time satisfies the Vogel-Fulcher (VF) law.[3,13] It allows us to replace τ in Eq. (10) by

$$\tau = \tau_0 e^{U/(T-T_0)} \tag{24}$$

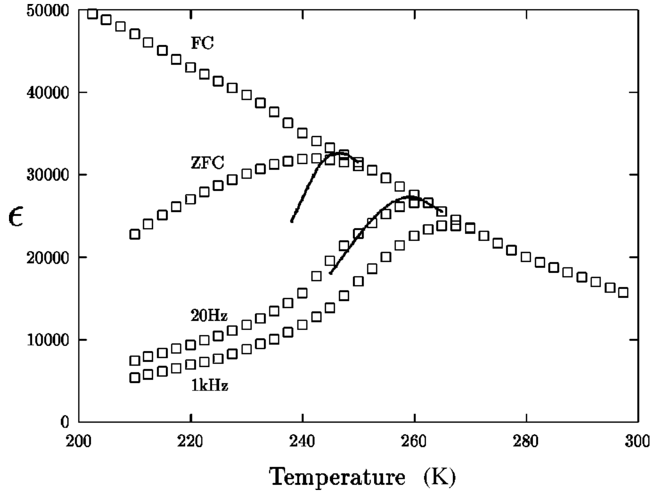

FIG. 2. Experimental (squares) and reconstructed (solid lines) values of the dielectric permittivity in PMN. Experimental values are reproduced from Ref. 13.

and introduces the temperature-independent distribution function $\tilde{g}(U)$ of potential barriers.[20,17] We obtain ($t > \tau_0$)

$$q(t) = 1 - \int_0^{(T-T_0)\ln(t/\tau_0)} dU\, \tilde{g}(U). \tag{25}$$

Thus, q is a function of $(T-T_0)\ln(t/\tau_0)$ that leads to the validity of the scaling relation $q(t,T) = q(t_1, T_1)$ with

$$T_1 = (T - T_0)\frac{\ln\left(\frac{t}{\tau_0}\right)}{\ln\left(\frac{t_1}{\tau_0}\right)} + T_0. \tag{26}$$

Using Eqs. (26), (14), and (16), one can reconstruct the values of $q(t,T)$ and $Q'(\omega,T)$ and, therefore, the values of $\epsilon(\omega)$ by employing the experimental data on $\epsilon(\omega_1)$ at a given frequency ω_1.

The reconstructed values of $\epsilon(\omega)$ at $\omega = 20$ Hz are presented in Fig. 2 (solid line), where the data at $\omega_1 = 1$ kHz have been taken as a reference. The parameters of the fit are $T_0 \approx 223$ K and $\tau_0 \approx 4 \times 10^{-11}$ sec, which are almost the same as obtained in Ref. 13, $T_0 \approx 224$ K and $\tau_0 \approx 4.3 \times 10^{-11}$ sec. One can see that at high temperatures ($T > 245$ K) the reconstruction reproduces the experimental data with good accuracy. The deviation between the reconstructed and experimental data for lower temperatures is due to violation of the VF law in PMN already discussed in Refs. 3 and 13. The self-consistency of the model has been tested also by reconstructing the values of the zero-field-cooled permittivity using Eq. (20) and the same reference data as above. In order to find the time t entering Eq. (20), we use the fact that in the experiment[13] the dielectric constant was measured by slowly heating the crystal from 80 K at the rate 0.5 K/min. It gives, e.g., the estimate $t = 330$ min at $T = 245$ K. The result of the reconstruction is shown in Fig. 2 by the solid line. As one can see from Fig. 2, the position and the height of the maximum of ϵ_{ZFC} are reproduced with very good accuracy. The deviation between the reconstructed values and experimental data is due to violation of the VF law taking place at low temperatures. Indeed, the temperature of the maximum of ϵ_{ZFC} corresponds to $T \approx 245$ K, which, as discussed above, is the boundary value for the validity of the VF law. If the VF law would be valid for all temperatures then both ϵ_{ZFC} and $\epsilon(\omega)$ would approach the value ϵ_0 at $T = T_0$. One can see that this tendency is reproduced by the interpolation of the solid curves in Fig. 2 to lower temperatures, thus clarifying the origin of the observed low-temperature deviations between the reconstructed and experimental values for ϵ_{ZFC}.

VI. CONCLUSION

The high dielectric constant of relaxor ferroelectrics signals that these materials are close to ferroelectric instability, which manifests itself in the critical slowing down of dynamics. In disordered relaxor materials, the critical slowing down is accompanied by glasslike freezing or cluster dynamics, which could be characterized by the nonequilibrium spin-glass order parameter. We have developed a dynamical model of the dielectric response in relaxors that explicitly takes into account the coexistence of critical and cluster dynamics and allows for separating them from the experimental observation.

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