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Polarization dynamics and formation of polar nanoregions in relaxor ferroelectrics

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Polarization dynamics and formation of polar nanoregions (PNR) in relaxor ferroelectrics is considered within a model of interacting short range polar clusters formed by off-center ions in highly polarizable materials. The model is applicable on the mesoscopic level and takes explicitly into account the distribution of cluster relaxation times and the existence of quenched random fields which control the size of PNR. Using self-consistent random field theory and continuous time random walk approximation, a relationship is established between the average polarization dynamics contributing to the low frequency dielectric response, and the local polarization dynamics determining NMR spin lattice relaxation time [Blinc *et al.*, Phys. Rev. B **63**, 024104 (2001); Cordero *et al.*, *ibid.* **71**, 094112 (2005)]. The lengthscale of PNR estimated from the obtained universal relationship between the parameters of the soft mode dispersion curve and the static dielectric permittivity is in agreement with the experiment [Vakhrushev *et al.*, Physica B **156-157**, 90 (1989)]. The predicted proportionality, between the intensity of diffuse neutron scattering from dynamic PNR and square of PNR correlation length, which has been recently a subject of controversy, is also in agreement with experiment by Vakhrushev *et al.* Therefore, we conclude that the model captures essential features of the static and dynamic behavior of relaxor ferroelectrics, and could be used for the characterization of new relaxor materials.

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I. INTRODUCTION

Relaxor ferroelectrics such as $PbMn_{1/3}Nb_{2/3}O_3$ (PMN), $PbSc_{1/2}Ta_{1/2}O_3$ (PST), or La-modified $PbZr_{1-x}Ti_xO_3$ (PLZT) have been a long standing puzzle for the ferroelectric community. Due to their unusual characteristics, very promising for industrial applications, numerous efforts have been spent recently to reveal the underlying physics of relaxor behavior. The main complexity includes compositional disorder in multivalent systems accompanied by high lattice polarizability. As a result, at low temperatures relaxor ferroelectrics transform into the new state of matter, intermediate between spin (dipole) glass and ordinary ferroelectrics, with well defined regions of correlated local polarization extended on the nanometer scale. It is now commonly accepted that the polar nanoregions (PNR) are responsible for extraordinary relaxor properties such as saturation of the field cooled dielectric permittivity, history dependent and hysteresis phenomena, large piezoelectric effect, etc.

There is now more and more evidences that unusual behavior of relaxor ferroelectrics is accompanied by off-center displacement of atoms from their centrosymmetrical positions. The role of the off-center ions in stimulating "glassy" properties of lead based relaxors was indicated first by Burns and Dacol,[1] who discovered the condensation of local polarization below well defined temperature T_d (Ref. 2) and suggested the applicability to these materials of the model[3] developed earlier for mixed perovskites like $KTa_{1-x}Nb_xO_3$ (KTN), or $K_{1-x}Li_xTaO_3$ (KLT), where off-center Li and Nb ions have been well identified[4] (also see recent review[5] and Ref. 6).

According to the model in Ref. 3, the local polarization in highly polarizable materials appears due to interaction of off-center ions with the lattice displacements associated with the soft polar mode. Such interaction leads to the crossover from the displacive type of polarization behavior to order-disorder type, characterized by the appearance of the dynamic central peak[7] in Raman and neutron scattering. Recent experiments (e.g., Refs. 8–12) confirm the appearance of dynamic central peak at $T<T_d$.

In the central peak regime off-center ions form short range reorienting clusters with the size $r_c>a$ (a is the lattice constant). Each cluster involves simultaneous displacements of other "host" atoms adjacent to a given off-center ion. Therefore effective dipole moment d^* of each cluster exceeds significantly the proper off-center ion dipole moment (defined as a product of the effective charge and off-center displacement), and the relaxor behavior can be thought to arise from the mutual interactions of such giant dipole moments accompanied by the effect of compositional disorder.

A consequence of the compositional disorder is the existence of quenched random field resulting in the suppression of ferroelectric phase transition due to strong local field fluctuations. It has been shown[13,14] that the effect of local field fluctuations could be included in a self-consistent manner, similar to Curie-Weiss approach (but avoiding mean field approximation), if one introduces the random field probability density. Such self-consistent random field theory (SCRFT) has been already applied to interacting polar clusters in PMN and PST to explain anomalies of dielectric permittivity, the crossover from relaxor to ferroelectric phase, E-T phase diagram, etc.[15]

In the present paper we concentrate on the correlation effects between short range polar clusters resulting in the formation of PNR. The latter are the regions of correlated polarization whose correlation length could exceed significantly the cluster size r_c. The theory developed below describes the features of a single "average" polar nanoregion, size of which is equal to the average correlation length $R_c \gg r_c$. Therefore the possible interactions between different PNR are not taken into consideration. Also we consider

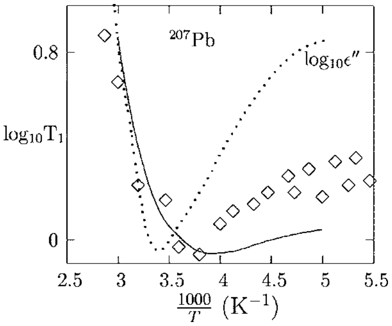

FIG. 1. Experimental (squares) and theoretical (solid line) temperature dependence of ^{207}Pb NMR spin lattice relaxation time T_1 in PMN at Larmor frequency 79.4 MHz. Experimental data are taken from Ref. 20. Theoretical curve is calculated from Eq. (44). Dotted line reproduces the temperature dependence of $\log_{10}\epsilon''(\omega)$ at $\omega = 112$ MHz taken from Ref. 26. Vertical positions of the dotted and solid lines are adjustable parameters.

transverse optic-type lattice displacements of the soft mode symmetry,[16] which contribute to the formation of "dynamic PNR" responsible for the finite width of the Raman central peak and large dielectric response of relaxors. We therefore will not discuss the origin of quasistatic PNR persisting at lower temperatures.[17–19]

In Sec. III we calculate the static structural factor of polarization and estimate the lengthscale of PNR. In Sec. IV we consider dynamic effects extending approach[15] on the account of fluctuation dynamics. A complexity of polarization dynamics in relaxor ferroelectrics stems from the coexistence of the critical dynamics, a common feature of any pretransitional phenomena, and the glassy dynamics of polar clusters. The effect of such dichotomy on the dielectric response, which is a probe of long wave length polar fluctuations, was analyzed earlier.[15] Here we will provide more emphasis on the local polarization dynamics which control, e.g., NMR spin lattice relaxation time T_1.[20,21]

Interrelation between the dynamics of long wavelength fluctuations (contributing to the dielectric response) and the local polarization dynamics provides an important test for distinguishing between the different models of relaxor behavior. A widely used model considers each PNR as an independent relaxor characterized by a single exponential dielectric relaxation. It implies that the observable experimental characteristics are the average values over the different PNR.[22–26] With the additional assumption that a PNR fluctuates as a whole, i.e., that the average and local polarization obey the same relaxation law, one comes to the conclusion[21,27] that T_1^{-1} should be proportional to the imaginary part of the dielectric susceptibility measured at NMR frequency.

However, this scenario is not realized in PMN (see Fig. 1) and apparently in other relaxors as well.[21] The temperature slope of ϵ'' is much more sharp than the slope T_1^{-1}, and the position of the maximum of T_1^{-1} and ϵ'' do not necessarily coincide.

A possible explanation of this inconsistency is that the local polar clusters inside PNR are not tight binding entities, and the local polarization could propagate along the PNR by diffusion type modes similarly to that in regular ferroelectrics. At the temperatures where dynamics of the average polarization undergoes critical slowing down, the diffusion type relaxation of local polarization dominates and partially smooths the effect of the critical slowing down on the local polarization dynamics.

In Sec. IV we will show that the effect of the distribution of cluster relaxation times on polarization dynamics could be described in terms of continuous time random walk stochastic process which has been widely explored for the analysis of different transport phenomena and aging effects in disordered solids. In Sec. V the predictions of the theory are compared with the experiment in PMN.

II. MODEL

We start from the assumption that in relaxor ferroelectrics due to compositional disorder some atoms might occupy off-center positions near the vacant lattice sites and form short range polar clusters involving displacements of adjacent atoms. Orientation of each off-center ion could be described by the unit vector $\mathbf{l}_i$ located at the ith unit cell characterized by the radius vector $\mathbf{r}_i$, and their interaction energy could be written in the form

$$V = -\frac{1}{2}\sum_{i,j=1}^{N}{}' \mathbf{l}_i \cdot \hat{\mathbf{J}}_{ij} \cdot \mathbf{l}_j. \tag{1}$$

Σ_j' means the summation over the unit cells occupied by off-center ions. We assume that off-center ions are randomly distributed over the unit cells with the average atomic concentration c, neglecting the correlation between the chemical and polar ordering. $\hat{\mathbf{J}}_{ij} \equiv \hat{\mathbf{J}}_{ij}(\mathbf{r}_{ij})$ with $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$; N is the total number of off-center ions in the crystal.

According to the model in Ref. 3, Fourier components $\mathbf{J}_q$ of the interaction matrix $\mathbf{J}_{ij} = \Sigma_q \mathbf{J}_q e^{i\mathbf{q}\cdot\mathbf{r}_{ij}}$ (where Σ_q corresponds to the summation over the reciprocal regular lattice) could be written as

$$\mathbf{J}_q = \frac{4\pi d^{*2}}{\epsilon_l}\mathbf{K}_q, \quad \mathbf{K}_q = \frac{\mathbf{g}_q}{V_0(1 + r_c^2 q^2)}, \tag{2}$$

with $\hat{\mathbf{g}}_q$ standing for a projection operator of transverse polarization fluctuations ($\mathbf{g}_q^2 = \mathbf{g}_q$)

$$\hat{g}_{\alpha\beta} = \left(\delta_{\alpha\beta} - \frac{q_\alpha q_\beta}{q^2}\right), \tag{3}$$

$\alpha, \beta = x, y, z$, $\delta_{\alpha\beta}$ is a Kronecker δ symbol, V_0 is the crystal volume, and ϵ_l is the "lattice" dielectric susceptibility at frequencies higher than the typical reorientation frequencies of off-center ions.

The form (2) of J_q assumes that the interaction J_{ij} is mediated by the high frequency polar lattice fluctuations. These fluctuations could be either in the form of soft phonon mode or a highly polarizable relaxational mode contributing to the central peak.[28] The only requirement is that off center ion reorientations are slow enough to ensure the validity of the adiabatic approximation: lattice polarization induced by off-center ions adiabatically follows the instantaneous off-center ion orientation. This induced polarization could be written as[3]

$$\mathbf{P}(\mathbf{r}_i) = d^* \sum_j{}' \mathbf{K}_{ij}(\mathbf{r}_{ij}) \cdot \mathbf{l}_j. \tag{4}$$

The meaning of Eqs. (4) and (2) is that each off-center ion polarizes the lattice near itself by the displacement of other atoms forming a short range cluster of the size r_c $[\int d\Omega K(\mathbf{r}) \sim \exp(-r/r_c)]$. Thus, Eq. (1) could be interpreted as the energy of the cluster-cluster interactions.

The first isotropic term in Eqs. (2) and (3) is responsible for ferroelectric correlations between cluster giant dipole moments and is significant if r_c exceeds the lattice constant. The second term describes the dipolar anisotropy competing with ferroelectric ordering.

Quenched random field $\mathbf{E}_i^{\mathrm{QF}}$ originated from compositional fluctuations in multivalent systems[29–31] is an additional source of anisotropy preventing the ferroelectric ordering. It is commonly accepted nowadays that in relaxors this source of anisotropy is much stronger than dipolar anisotropy. Thus, the resulting Hamiltonian characterizing the cooperative behavior of polar clusters in relaxor ferroelectrics could be written in the form

$$H = \sum_i{}' \mathbf{l}_i \cdot \mathbf{E}_i, \tag{5}$$

where $\mathbf{E}_i$ is the local field (in energy units) which experiences ith polar cluster:

$$\mathbf{E}_i = \sum_j c_j \hat{\mathbf{J}}_{ij} \cdot \mathbf{l}_j + \mathbf{E}_i^{\mathrm{QF}} + \mathbf{E}_{\mathrm{ex}}, \tag{6}$$

$E_{\mathrm{ex}} = d^* \mathcal{E}_{\mathrm{ex}}$, where $\mathcal{E}_{\mathrm{ex}}$ is the applied electric field, Σ_i denotes the summation over all unit cells, and c_i is the random occupation number of the unit cell ($c_i = 0, 1$).

Note that in contrast with the model[32] the present approach uses short range interaction potential instead of infinite range potential. This allows to estimate the correlation effects which is the main goal of the present paper. Also the model is not restricted to the constraint of continuous symmetry of the vectors $\{l_i\}$, and could be applied to relaxors with cubic and uniaxial[33] symmetry as well.

III. SELF-CONSISTENT RANDOM FIELD THEORY FOR STATIC POLAR CORRELATIONS

According to Eq. (4), static structure factor of polarization $\hat{\mathbf{S}}_q = \langle \mathbf{P}_q^2 \rangle$ could be written as

$$\hat{\mathbf{S}}_q = \mathrm{Tr}(\hat{\mathbf{K}}_q \cdot \hat{\mathbf{C}}_q \cdot \hat{\mathbf{K}}_q), \tag{7}$$

where $\hat{\mathbf{C}}_q$ is the static structure factor of off-center ion orientations

$$\hat{C}_q^{\alpha\beta} = n\left(\langle l_{i\alpha} l_{i\beta} \rangle + \sum_{j \neq i} \langle l_{i\alpha} l_{j\beta} \rangle_q \right). \tag{8}$$

A traditional approach to calculate the structure factor is the use of the mean field theory along with the fluctuation-dissipation theorem.[34] This approach is not applicable in relaxors due to strong local field fluctuations. SCRFT has been developed[13,14] as an alternative to mean field theory to take into account the probability density of the local field (instead of only mean field parameter in Curie-Weiss theory). Before considering the correlation effects, we review briefly the general framework of the SCRFT and introduce the notations.

A. Long range order parameter and static field cooled susceptibility

The average polarization induced by off-center ions could be written as

$$\langle \mathbf{P} \rangle = n d^* \mathbf{L}, \tag{9}$$

where $n = N/V_0$ and $\mathbf{L} = \frac{1}{N} \Sigma_i \langle \mathbf{l}_i \rangle$ is the orientational order parameter. The SCRFT equation for long range orientational order parameter $\mathbf{L}$ reads

$$\mathbf{L} = \int d\mathbf{E} \langle \mathbf{l} \rangle_E f(\mathbf{E}, \mathbf{L}), \tag{10}$$

where $\langle \mathbf{l} \rangle_E$ characterizes the thermal average value of the vector $\mathbf{l}_i$ in the local field $\mathbf{E}$ and $f(\mathbf{E}, \mathbf{L})$ is the probability density of the local field depending parametrically on the value of $\mathbf{L}$

$$f(\mathbf{E},\mathbf{L}) = \int d\mathbf{E}_i^{\mathrm{QF}} f(\mathbf{E}_i^{\mathrm{QF}}) \overline{\left\langle \delta\left(\mathbf{E} - \sum_j c_j \hat{\mathbf{J}}_{ij} \mathbf{l}_j - \mathbf{E}_i^{\mathrm{QF}} - \mathbf{E}_{\mathrm{ex}} \right) \right\rangle_{\{l_i\}}}. \tag{11}$$

Here $\langle \ldots \rangle_{\{l_i\}}$ denotes the thermal average over the variables $\{\mathbf{l}_l\}$, and the overbar stands for configurational average over occupation numbers $\{c_i\}$; $f(\mathbf{E}^{QF})$ is the probability density of quenched random field.

The dependence $f(\mathbf{E}, \mathbf{L})$ on $\mathbf{L}$, a key point of SCRFT, makes Eq. (10) a self-consistent equation with respect to $\mathbf{L}$ which could be considered as a generalization of the Curie-Weiss mean field equations on the account of local field fluctuations. Indeed, in the absence of fluctuations one could replace $f(\mathbf{E}, \mathbf{L})$ by $\delta(\mathbf{E} - E_0 \mathbf{L})$ and Eq. (10) reduces to the usual Curie-Weiss mean field equation (δ is the Dirac delta function, and $E_0 = 4\pi n d^{*2}/\epsilon_l$ is a mean field parameter). However, when the local field fluctuations are significant, SCRFT could lead to very different results compared with Curie-Weiss theory. In particular, it reproduces a crossover from long range to mesoscopic order depending on the strength of the local field fluctuations thus providing, e.g., a natural explanation of relaxor to ferroelectric phase transitions with a slight change of system composition.

Note also that Eq. (10) is exact for two and eight orientations of the vectors $\{l_i\}$, as soon as the function $f(\mathbf{E}, \mathbf{L})$ is known, and could be considered as a reasonable approximation for the other models of the off-center ion energy surface. A remark should be made here regarding spherically symmetrical distribution of off-center ion displacements which relates such relaxors to the class of systems with continuous symmetry. As known, in this case both dipole anisotropy[35] and quenched random fields[36] are destroying factors of long range order due to transverse Golstone mode type fluctuations of the local field (not included in the formulation of SCRFT). However, in relaxors, in contrast with magnetic Heisenberg systems, the local barriers for cluster reorientations suppress the Golstone type fluctuations thus imposing the limitations on continuous symmetry approximation.

The function $f(\mathbf{E},\mathbf{L})$ depends on fluctuations of the potential $\hat{\mathbf{J}}(r_{ij})$ due to fluctuations of occupation numbers $\{c_i\}$ (so called random bond fluctuations considered in detail in Ref. 14); on the thermal fluctuations of vectors $\{l_i\}$; and on the probability density of the quenched random fields $f^{\mathrm{QF}}(\mathbf{E})$.[37,13] Apparently in relaxor ferroelectrics quenched random fields is the main source of local field fluctuations.[29,30,38] In this case the problem is simplified, since we could neglect the thermal fluctuations of $\{\mathbf{l}_i\}$ and the function $f(\mathbf{E},\mathbf{L})$ reduces to

$$f(\mathbf{E},\mathbf{L}) = f^{\mathrm{QF}}(\mathbf{E} - E_0\mathbf{L} - \mathbf{E}_{\mathrm{ex}}). \tag{12}$$

Equations (10) and (12) could be transformed to modified Landau type free energy, and has been applied earlier[13] to PMN and PST to describe the crossover from relaxor to ferroelectric state, *E*-*T* phase diagram, first order phase transitions, etc.

For example, assuming that the external field is parallel to the *z* axis, static or field cooled permittivity could be calculated using its definition $\epsilon_{\mathrm{FC}} - \epsilon_l = 4\pi\langle P_z\rangle/\mathcal{E}_{\mathrm{ex}}$ at $\mathcal{E}_{\mathrm{ex}} \to 0$ resulting in

$$\epsilon_{\mathrm{FC}} - \epsilon_l = \frac{\epsilon_l k(T)}{1 - k(T)}, \tag{13}$$

where

$$k(T) = E_0 \int dE_z \frac{\partial\langle l_z\rangle_E}{\partial E_z} f^{\mathrm{QF}}\,(E_z)|_{L=L_s}. \tag{14}$$

L_s is the equilibrium order parameter being a solution of Eq. (10) below the ferroelectric transition temperature T_c. For a second order phase transition $k(T_c)=1$ [in mean field approximation $k(T)=T_c/T$, where $T_c=E_0/3$ for three-dimensional vectors $\{\mathbf{l}_i\}$]. In relaxors with incipient ferroelectric order like PMN, $k(T)<1$ and $L_s=0$ at any temperature. In this case Eq. (13) reproduces the low temperature saturation of the field cooled permittivity, which in disordered systems deviates from the true equilibrium permittivity.

At $T \to 0 k(T) \propto f^{\mathrm{QF}}(E=0)$ meaning that the dielectric response is proportional to the fraction of clusters not frozen in random fields. This is a well known effect of static freezing in quenched random field. The dynamic freezing at finite frequencies will be discussed in Sec. III.

B. Static structure factor of polar correlations: Formation of PNR

The structure factor $\hat{\mathbf{C}}_q$ could be derived within SCRFT using the following steps. First derive the self-consistent linear equations for the local variables $\{\langle l_i\rangle\}$ assuming $E_{\mathrm{ex}}=0$

$$\langle l_i\rangle = \int d\mathbf{E}_i^{\mathrm{QF}} f(\mathbf{E}_i^{\mathrm{QF}}) \left\langle \delta\left(\mathbf{E} - \sum_j c_j \hat{\mathbf{J}}_{ij}\cdot\mathbf{l}_j - \mathbf{E}_i^{\mathrm{QF}} - \mathbf{E}_{\mathrm{ex}}\right)\right\rangle, \tag{15}$$

where $\langle\cdots\rangle_{\mathrm{QF}}$ denotes the average over the quenched random field. In contrast with Eq. (10) we do not take here the average over the site occupation numbers $\{c_i\}$.

Considering as above quenched random fields as the main source of local field fluctuations, i.e., neglecting the thermal fluctuations of variables $\{\mathbf{l}_i\}$, we could write the following equation instead of Eqs. (10) and (12):

$$\langle l_{i\alpha}\rangle = \int dE_\alpha \langle l_\alpha\rangle_E f^{\mathrm{QF}}\left(E_\alpha - \sum_j c_j J_{ij}^{\alpha\beta}\langle l_{j\beta}\rangle\right) \tag{16}$$

or, in the linear approximation

$$\langle l_{i\alpha}\rangle = \sum_j c_j \Phi_{ij}^{\alpha\beta}\langle l_{j\beta}\rangle, \tag{17}$$

where

$$\hat{\Phi}_{ij} = \frac{k(T)}{E_0}\hat{\mathbf{J}}_{ij}. \tag{18}$$

$\hat{\Phi}_{ij}$ could be considered as the effective interaction energy, renormalized by the local field fluctuations. In the mean field approximation $\hat{\Phi}_{ij}=\hat{\mathbf{J}}_{ij}/T$.

Note that the validity of approximation (16) depends on the amplitude of quenched random field fluctuations. An alternative approach which takes into account the thermal fluctuations of the variables $\{\mathbf{l}_i\}$ has been considered in Ref. 14.

Following the Ornstein-Zernike approach (random phase approximation), assume that the vector l_0 in Eq. (17) has a given (random) orientation and use the definition of the correlation function

$$\hat{G}_{i0}^{\alpha\beta} \equiv \langle l_{i\alpha} l_{0\beta}\rangle = \langle\langle l_{i\alpha}\rangle_{l_0} l_{0\beta}\rangle, \tag{19}$$

where $\hat{\mathbf{G}}_{ij}=\hat{\mathbf{G}}(\mathbf{r}_{ij})$ and $\langle\mathbf{l}_i\rangle_{l_0}$ is the conditional average value of $\mathbf{l}_i$ at given value of $\mathbf{l}_0$. The equation for $\hat{\mathbf{G}}_{i0}$ could then be obtained by multiplying Eq. (17) by $l_{0\beta}$ and taking the average. Thus we have

$$\hat{\mathbf{G}}_{i0} = c\sum_j \hat{\Phi}_{ij}\hat{\mathbf{G}}_{j0} + \frac{1}{3}\hat{\Phi}_{i0}. \tag{20}$$

Taking the Fourier transform of Eq. (20), we obtain

$$\hat{\mathbf{G}}_q = \frac{1}{3}\frac{\hat{\Phi}_q}{1 - N\Phi_q}, \tag{21}$$

which with the use of Eqs. (7) and (13) gives the final form for the static structure factor of transverse polarization (assuming $\epsilon_{\mathrm{FC}} \gg \epsilon_l$)

$$\hat{\mathbf{S}}_q = \frac{nd^{*2}}{3\epsilon_l}\frac{\epsilon_{\mathrm{FC}}}{(1+R_c^2q^2)(1+r_c^2q^2)}\hat{\mathbf{g}}, \tag{22}$$

where

$$R_c^2 = \frac{\epsilon_{\mathrm{FC}}}{\epsilon_l} r_c^2. \qquad (23)$$

Equations (22) and (23) show that the correlation effects between short range polar clusters result in the formation of PNR. The latter are the regions of correlated polarization whose correlation length R_c exceeds significantly the cluster size r_c at temperatures where $\epsilon_{\mathrm{FC}} \gg \epsilon_l$. One can see from Eq. (23) that the lengthscale of PNR could be easily estimated from the rather universal relationship between the parameters ϵ_{FC}, ϵ_l, and r_c.

The correlation function (22) determines the energy integrated scattering intensity $I(q) \propto S_q$ measured in x-ray or diffuse neutron scattering experiments. One can see from Eq. (22) that $I(q=0) \propto \epsilon_{\mathrm{FC}}(T)$, and $I(q)$ is a Lorentzian function at $q^2 r_c^2 \ll 1$.

Note that in Ref. 39 the expression for S_q similar to Eq. (22) has been obtained with the use of the fluctuation-dissipation theorem and conventional mean field theory providing

$$\epsilon_{\mathrm{FC}} = \frac{\epsilon_l}{1 - T_c/T}, \quad T_c = \frac{4\pi n d^{*2}}{3\epsilon_l},$$

as discussed in Sec. III A.

However, the effect of the local field fluctuations on the structure factor cannot be treated with the use of the fluctuation-dissipation theorem, which is a well known consequence of the configurational disorder.[40]

IV. POLARIZATION DYNAMICS AS CONTINUOUS TIME RANDOM WALK

A. Basic equations

In order to describe the polarization dynamics of PNR, we start from the traditional assumption that nonequilibrium systems approach equilibrium via the quasiequilibrium states (or states of local equilibrium). In our case this assumption corresponds to the validity of the following master equation:

$$\frac{d\langle \mathbf{l}_i(t)\rangle}{dt} = -\frac{1}{\tau_i}[\langle \mathbf{l}_i(t)\rangle - \langle \tilde{\mathbf{l}}_i(t)\rangle], \qquad (24)$$

where τ_i is the relaxation time of the dipole moment of ith cluster, and $\tilde{\mathbf{l}}_i(t)$ is the local equilibrium value of the vector $\mathbf{l}_i$ satisfying the equation (assuming $E_{\mathrm{ex}}=0$)

$$\langle \mathbf{l}_i(t)\rangle = \sum_j c_j \hat{\Phi}_{ij} \cdot \langle \mathbf{l}_j(t)\rangle. \qquad (25)$$

Equations (24) and (25) could be combined as

$$\frac{d\langle \mathbf{l}_i(t)\rangle}{dt} = -\frac{1}{\tau_i}\left(\langle \mathbf{l}_i(t)\rangle - \sum_j c_j \hat{\Phi}_{ij} \cdot \langle \mathbf{l}_j(t)\rangle\right). \qquad (26)$$

Due to the compositional disorder the relaxation time τ_i is a random variable characterized by the probability density $g(\tau_i)$. The distribution of relaxation times leads to the nonexponential relaxation, which is described by the relaxation function

$$q(t) = \langle e^{-t/\tau_i}\rangle_{\tau_i} = \int_{\tau_{\min}}^{\tau_{\max}} d\tau_i g(\tau_i) e^{-t/\tau_i}, \qquad (27)$$

where $\langle \ldots \rangle_{\tau_i}$ denotes the average over the distribution of relaxation times.

For the smooth functions $g(\tau_i)$, Eq. (27) reduces to

$$q(t) \approx \int_t^{\tau_{\max}} d\tau_i g(\tau_i). \qquad (28)$$

Thus, the variable $q(t)$ describes the fraction of clusters effectively frozen at time t, and therefore has a meaning of spin glass order parameter at finite time scale.[15]

In order to average Eq. (26) over the distribution of relaxation times it is convenient to rewrite it in the equivalent integral form

$$\langle \mathbf{l}_i(t)\rangle = \mathbf{l}_i(0) e^{-t/\tau_i} + \frac{1}{\tau_i}\int_0^t dt' e^{-t'/\tau_i} \sum_j c_j \hat{\Phi}_{ij} \langle \mathbf{l}_j(t-t')\rangle. \qquad (29)$$

When averaging over the distribution of relaxation times we used the following decoupling approximation in Eq. (29)

$$\left\langle \frac{1}{\tau_i} e^{-t'/\tau_i} \langle \mathbf{l}_j(t-t')\rangle \right\rangle_{\{\tau_i\}} = \left\langle \frac{1}{\tau_i} e^{-t'/\tau_i} \right\rangle_{\tau_i} \langle\langle \mathbf{l}_j(t-t')\rangle\rangle_{\{\tau_i\}}. \qquad (30)$$

One can show by iterations that decoupling approximation (30) corresponds to neglecting correlations between τ_i, τ_j for $i \neq j$.

With the use of approximation (30), Eq. (29) reduces to the form

$$\langle l_i(t)\rangle = l_i(0) q(t) - c\int_0^t dt' \frac{dq(t')}{dt'} \sum_j \Phi_{ij} \langle l_j(t-t')\rangle, \qquad (31)$$

where we replaced the occupation numbers c_j by their average value, and for simplicity of notations included in the symbol $\langle \ldots \rangle$ the average over the relaxation times as well.

Equation (31) has a well defined stochastic interpretation. It describes polarization fluctuations as continuous time random walk stochastic process (with dissipation), which has been widely used to describe different kinetic phenomena in systems with configurational disorder, such as hopping conductivity,[41] spin diffusion,[42] transport of noncoherent excitons,[43] etc. Recently this approach has been extended to describing aging effects in disordered systems (see, e.g., Ref. 44). Decoupling approximation of the type of Eq. (29) has been introduced in Refs. 42 and 43.

Equation (31) is the basic equation for the analysis of dynamic correlation and autocorrelation (or local correlation) effects. We will consider them separately.

B. Dynamic structure factor of polarization

In analogy with Eqs. (7) and (8), the dynamic structure factor of polarization

$$\hat{\mathbf{S}}_{q\omega} = \langle \mathbf{P}_q(t) \mathbf{P}_q(0)\rangle_\omega \qquad (32)$$

is defined as

$$\hat{\mathbf{S}}_{q\omega} = \mathrm{Tr}(\hat{\mathbf{K}}_q \cdot \hat{\mathbf{C}}_{q\omega} \cdot \hat{\mathbf{K}}_q), \tag{33}$$

where

$$\hat{\mathbf{C}}_{q\omega} = n[\langle \mathbf{l}_i(t)\mathbf{l}_i(0)\rangle + N\langle \mathbf{l}_i(t)\mathbf{l}_j(0)\rangle_{q\omega}]. \tag{34}$$

It is convenient to present $\hat{\mathbf{C}}_{q\omega}$ in terms of Fourier components $\mathbf{l}_q = \Sigma_i \mathbf{l}_i e^{-\mathbf{qr}_i}$ as

$$\hat{\mathbf{C}}_{q\omega} = \int_{-\infty}^{\infty} dt e^{-i\omega t}\hat{\mathbf{C}}_q(t), \quad \hat{\mathbf{C}}_q(t) = \langle \mathbf{l}_q(t)\mathbf{l}_q(0)\rangle. \tag{35}$$

Taking into account that $\langle \mathbf{l}_q(t)\mathbf{l}_q(0)\rangle = \langle\langle \mathbf{l}_q(t)\rangle_{l_q(0)}\mathbf{l}_q(0)\rangle$, where $\langle \mathbf{l}_q(t)\rangle_{l_q(0)}$ is the average of $\mathbf{l}_q(t)$ under the condition that $\langle \mathbf{l}_q(t=0)\rangle = \mathbf{l}_q(0)$, we obtain the equation for $\hat{\mathbf{S}}_q(t)$ using the spatial Fourier transform of Eq. (31)

$$\hat{\mathbf{S}}_q(t) = \hat{\mathbf{S}}_q q(t) - \int_0^t dt' \frac{dq(t')}{dt'} N\hat{\Phi}_q \cdot \hat{\mathbf{S}}_q(t-t'), \tag{36}$$

where static structure factor $\hat{\mathbf{S}}_q$ is given by Eq. (22).

Solving Eq. (36) we obtain dynamic structure factor $\hat{\mathbf{S}}_{q\omega}$ in the form

$$\hat{\mathbf{S}}_{q\omega} = \mathrm{Im}\,\frac{\hat{\mathbf{S}}_q}{\omega}\frac{1-Q(\omega)}{1-N\Phi_q Q(\omega)}, \tag{37}$$

where Im stands for the imaginary part of the expression and

$$Q_\omega = \int_0^\infty dt e^{-i\omega t}\frac{dq}{dt} = \left\langle \frac{1}{1+i\omega\tau} \right\rangle \tau. \tag{38}$$

At $\omega \to 0$ we have $Q'_\omega \to 1$, $Q''_\omega \to \omega\langle\tau\rangle$, and Eq. (37) transforms to the Lorentzian form

$$\hat{\mathbf{S}}_{q\omega} = \hat{\mathbf{S}}_q \frac{\tau_q}{1+\omega^2\tau_q^2}, \quad \tau_q = \frac{\langle\tau\rangle}{1-N\Phi_q}, \tag{39}$$

where τ_q is the characteristic relaxation time for the qth Fourier component of polarization fluctuations. One can see that for $q=0$ the relaxation time $\tau_{q=0} = \frac{\langle\tau\rangle}{1-k(T)}$, meaning that the slowing down of relaxation with the temperature decrease consists of two contributions: (a) slowing down of cluster dynamics described by the dependence $\langle\tau(T)\rangle$ and (b) the critical slowing down described by the factor $[1-k(T)]^{-1}$.

Equation (39) is similar to that in regular order-disorder ferroelectrics, with relaxation time τ replaced by its average value $\langle\tau\rangle$ and T_c/T replaced by the function $k(T)$. The deviation from regular behavior is pronounced with the increase of the parameter $\omega\langle\tau\rangle$ when the dynamic structure factor deviates from the Lorentz form.

For the typical condition $1 - N\Phi_q Q'_\omega \gg N\Phi_q Q''_\omega$, or $1 - kQ'_\omega \gg kQ''_\omega$, providing that the real part of the frequency dependent dielectric permittivity exceeds its imaginary part [see Eq. (46)], we obtain from Eqs. (37) and (22)

$$\hat{\mathbf{S}}_{q\omega} = \frac{nd^{*2}}{3\epsilon_l}\frac{Q''}{\omega}\frac{\epsilon_{\mathrm{FC}}}{(1+R_c^2q^2)(1-kQ'_\omega+r_c^2q^2)}\hat{\mathbf{g}}. \tag{40}$$

Dynamic structure factor determines inelastic scattering intensity. It determines also the local correlation effects.

C. Local polar correlations

Fourier components of autocorrelation function of local polar fluctuations are equal to

$$G_\omega^{\mathrm{loc}} = \sum_q S_{q\omega}. \tag{41}$$

Since all modes with $q>0$ contribute to autocorrelation function (41), the effect of the critical slowing down on local dynamics is less pronounced than on the mode with $q=0$. Indeed, even at $k \to 1$ when $\tau_{q=0} \to \infty$, the relaxation times of the modes with $q>0$ have the finite values $\tau_q^{-1} = Dq^2$, due to diffusion like propagation of the local polarization ($D = r_c^2/\langle\tau\rangle$ is the diffusion coefficient).

The correlation time of local polarization fluctuations is given by

$$\tau_{\mathrm{loc}} = \frac{\sum_q S_{q\omega=0}}{\sum_q S_q}, \tag{42}$$

which with the account of Eqs. (21) and (39) results in

$$\tau_{\mathrm{loc}} = \frac{(\sqrt{1-k}+1)}{2\sqrt{1-k}}\langle\tau\rangle. \tag{43}$$

One can see that for the typical for relaxor values of the parameter $1-k \sim 10^{-2}$, the effect of the critical slowing down on the local polarization dynamics is significant.

For the correlation function G_ω^{loc} at finite frequencies we obtain with the use of Eqs. (40) and (41)

$$G_\omega^{\mathrm{loc}} \propto \frac{Q''_\omega}{\omega}\frac{1}{\sqrt{\epsilon'(\omega)^{-1}} + \sqrt{\epsilon_{\mathrm{FC}}^{-1}}}, \tag{44}$$

where $\epsilon'(\omega, T)$ is the real part of the complex dielectric susceptibility.

The correlation function G_ω^{loc} determines in particular NMR spin lattice relaxation rates

$$T_1^{-1} \propto G_{\Omega_{\mathrm{Larm}}}^{\mathrm{loc}}, \tag{45}$$

where Ω_{Larm} is the Larmor frequency. Therefore, Eq. (44) could be directly compared with the experiment.

When obtaining Eq. (44) we have taken into account that the relaxation function Q_ω determines as well the frequency dependent dielectric susceptibility. The dielectric susceptibility could be derived from Eq. (24) in the presence of applied electric field, or with the use of the simpler approach.[15] It has the form

$$\epsilon(\omega) - \epsilon_l = \frac{\epsilon_l k Q_\omega}{1-kQ_\omega}. \tag{46}$$

At $1-kQ'_\omega \gg kQ''_\omega$, which is equivalent of $\epsilon'' \gg \epsilon'$, we obtain

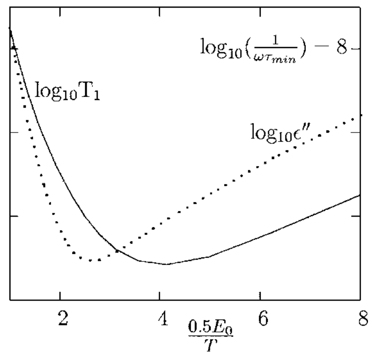

FIG. 2. Comparison of the theoretical temperature dependences of the NMR spin lattice relaxation time T_1 (solid line) and ϵ'' (dotted line) for the model of relaxor dielectric response presented in Fig. 3. Vertical position of the solid line is the adjustable parameter.

$$\epsilon'(\omega) = \frac{\epsilon_l}{1 - kQ'_\omega}, \tag{47}$$

$$\epsilon''(\omega) = \frac{\epsilon'(\omega)^2}{\epsilon_l} Q''_\omega. \tag{48}$$

One can see comparing Eqs. (44), (45), and (48) that although both T_1^{-1} and ϵ'' are proportional to Q''_ω, the temperature dependence of ϵ'' is enhanced by the strong temperature dependence of the multiplier $\epsilon'(\omega)^2$, compared with the rather weak temperature dependence of the multiplier

$$\frac{1}{\sqrt{\epsilon'(\omega)^{-1}} + \sqrt{\epsilon_{\mathrm{FC}}^{-1}}}$$

determining T_1. This result explains qualitatively the rather weak temperature dependence of NMR spin lattice relaxation time observed in PMN (Ref. 20) below the freezing temperature T_f (corresponding to the maximum of ϵ'').

In Fig. 2 we illustrate the capability of the theory in the description of local polarization dynamics comparing the behavior of $T_1(T)$ and $\epsilon''(T)$ for the model functions ϵ_{FC} and $Q_\omega(T)$, employed earlier[15] for demonstration of giant polydispersivity in dielectric response.

The temperature dependence of ϵ_{FC} used in the calculations is shown in Fig. 3. It reproduces the Curie-like high-temperature behavior of field-cooled permittivity and its high saturation value at low temperatures. Such T dependence of ϵ_{FC} could be obtained from Eqs. (13) and (14) with $\langle l_z \rangle_E = \frac{1}{\sqrt{3}} \tanh(E_z/\sqrt{3})$ (eight well potential for off-center ion reorientations), and Gaussian shape of $f(E)^{\mathrm{QF}}$ assuming that the width of $f(E)^{\mathrm{QF}}$ is slightly temperature dependent. The functions $Q'_\omega(T)$, $Q''_\omega(T)$, have been calculated assuming the Arrhenius temperature dependence of $\tau(T) = \tau_{\min} e^{U/T}$ and the typical shape of the reorientational barrier probability density $\tilde{g}(U)$ discussed in Ref. 15. The values of ϵ' and ϵ'' shown in Figs. 2 and 3 have been obtained with the use of Eqs. (47) and (48).

One can see from Fig. 2 that the temperature slope of T_1 is less sharp than the slope of ϵ''^{-1}, which indicates that the local polarization dynamics in relaxors is less sensitive to the critical slowing down than the dynamics of the average polarization. Also the positions of $T_1(T)$ minimum and ϵ'' maximum do not coincide, in qualitative agreement with the experiment.[20] More detailed comparison of the theory with the experiment will be discussed in the next section.

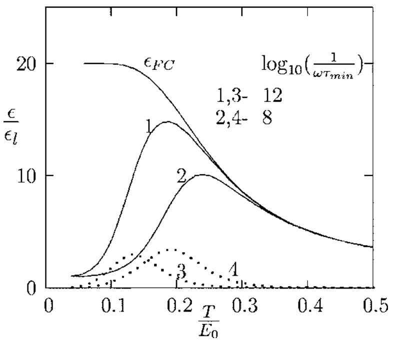

FIG. 3. Theoretical temperature dependences of the real (solid lines) and imaginary (dotted lines) parts of the dielectric permittivity for the different frequencies of applied field corresponding to the model functions $\epsilon_{\mathrm{FC}}(T)$ and $Q_\omega(T)$ discussed in the text.

V. COMPARISON WITH THE EXPERIMENT IN PMN

A. Two time scales associated with Nb and Pb reorientation dynamics

We presented above a physical model of formation and polarization dynamics of PNR in relaxor ferroelectrics. According to the model, finite size of polar nanoregions originates from the competition of indirect (mediated by high frequency polar lattice fluctuations) attractive interactions between off-center ions which favor the ferroelectric ordering, and the interaction of off-center ions with static random fields which induce the random anisotropy and prevent the ordering. The only prerequisites for the model to be applicable are (a) the existence of off-center displacements of atoms and their thermal reorientations between equivalent off-center positions and (b) high polarizability of the lattice at frequencies $\omega > \langle \tau \rangle^{-1}$.

The existence of off-center displacement of both Pb and Nb atoms in PMN are currently well documented.[1,20,19] It has been recognized also a particular role of Nb atoms in the formation and anomalies of the central peak observed in Raman scattering experiments.[10] These latest findings indicate that Nb atoms are responsible for the fast reorientation dynamics consistent with the fact that the multiwell potential for Nb reorientations is caused by the small Nb ionic radius. Apparently, Nb fast motion is responsible for the dielectric permittivity of PMN at high frequencies $\sim 10^{11}$ Hz. At these frequencies the dielectric susceptibility $\epsilon \sim \epsilon_{\mathrm{Nb}}$ is almost temperature independent. Its values, close to 1000 according to high frequency dielectric spectroscopy,[12] significantly exceed dielectric permittivity ϵ_{SM} corresponding to the soft mode [$\epsilon_{\mathrm{SM}} \sim 80$ at $T = 300$ K (Ref. 38)].

Off-center displacement of Pb has a very different origin. Apparently it is caused by the random electric fields due to charge unbalance inside a unit cell.[45] In this case one would not expect that for a given configuration of the random fields, the single Pb ion potential energy possesses multiminima structure. Pb reorientations would more probably take place if they associate with the motion of more than one Pb ion within clusters (i.e., the cluster potential energy is characterized by the multiwell structure). The time scale of such cluster dynamics would be much longer than the time scale of Nb dynamics, and responsible for the low frequency dielectric anomalies.

This picture reminds one in some sense, of a situation in $K_{1-x}Li_xTa_{1-y}Nb_yO_3$ with both Li and Nb ions being in off-center positions, such that fast Nb dynamics contributes to the Raman central peak[46] and slow Li dynamics contributes to low frequency dielectric dispersion.[47] In PMN (as well as in $K_{1-x}Li_xTa_{1-y}Nb_yO_3$) a crossover to central peak behavior (Nb relaxational mode) has been observed at high temperatures indicating the onset of strong interaction of off-center Nb ions with the soft mode polarization, which results in the indirect Nb-Nb interaction (1) and (2), where ϵ_l and r_c should be considered, respectively, as dielectric permittivity ϵ_{SM} and correlation length $r_{c_{\mathrm{SM}}}$ associated with the soft mode.[3] Indirect Nb-Nb interactions mediated by the soft mode lead to the enhancement of the Nb dielectric response ϵ_{Nb} and the associated correlation length $r_{c_{\mathrm{Nb}}}$ [according to Eq. (23), $r_{c_{\mathrm{Nb}}}^2=(\epsilon_{\mathrm{Nb}}/\epsilon_{\mathrm{SM}})r_{c_{\mathrm{SM}}}^2$].

When the temperature decreases and approaches freezing temperature T_f there becomes significant long wave length cooperative dynamics of short range clusters contributing to low frequency dielectric anomalies. This cooperative cluster dynamics originates from the indirect cluster-cluster interactions mediated by the polarization fluctuations associated with the Nb relaxational mode, since the latter is much more polarizable than the soft mode. Indirect cluster-cluster interaction is described by the same Hamiltonian (1) and (2), where now ϵ_l and r_c should be replaced, respectively, by high frequency Nb dielectric permittivity ϵ_{Nb} and the correlation length $r_{c_{\mathrm{Nb}}}$. The proposed picture of relaxor behavior of PMN allows one to make some quantitative predictions by comparing the theory with the experiment.

B. Length scale of polar nanoregion

According to Eq. (23) and the discussion above, the average size of PNR is given by

$$R_c^2 = \frac{\epsilon_{\mathrm{FC}}}{\epsilon_{\mathrm{Nb}}} r_{c_{\mathrm{Nb}}}^2 = \frac{\epsilon_{\mathrm{FC}}}{\epsilon_{\mathrm{SM}}} r_{c_{\mathrm{SM}}}^2 . \tag{49}$$

The parameter $r_{c_{\mathrm{SM}}}$ could be estimated from the soft mode dispersion curve $\omega_q^2=\omega_0^2+c^2q^2$, or $\omega_q^2=\omega_0^2(1+r_{c_{\mathrm{SM}}}^2q^2)$ with $r_{c_{\mathrm{SM}}}=\frac{c}{\omega_0}$. Using the neutron scattering data,[48] we obtain $r_{c_{\mathrm{SM}}}\approx 1.7a$ ($a\approx 4$ Å is the PMN lattice constant).

Since $\epsilon_{\mathrm{SM}}\propto\omega_0^{-2}$, the parameter $r_{c_{\mathrm{SM}}}^2/\epsilon_{\mathrm{SM}}$ is temperature independent. In this case Eq. (49) predicts the proportionality between the square of the correlation length R_c^2 and the static field cooled permittivity.

In order to demonstrate that this proportionality takes place in the experiment we plot in Fig. 4 the values of the inverse static dielectric susceptibility between 500 and 200 K (using the scaling coefficient). The experimental data are taken from Ref. 24 for $T<350$ K and from Ref. 49 for $T>350$ K (above $T>350$ the low frequency dielectric permittivity in PMN is frequency independent).

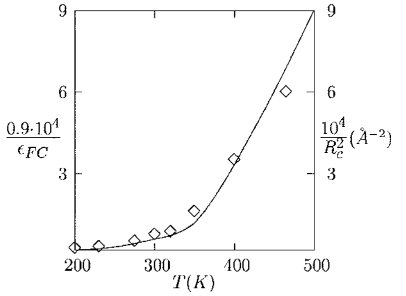

FIG. 4. Experimental temperature dependences of the inverse static dielectric permittivity (solid line) (Refs. 24 and 49) and square of the inverse correlation length of PNR in PMN (squares) (Ref. 22).

The values of R_c obtained in neutron scattering experiments[50,22] are presented on the same graph. Although, as follows from the analysis in Ref. 16, these values of R_c represent the combined effect of dynamic and static atom displacements (the latter are not under discussion in this paper), the contribution of dynamic displacements to R_c dominates at least at $T>250$ K, where the correlation length of static displacements is less than R_c.[18]

One can see that the proportionality $R_c^2\propto\epsilon_{\mathrm{FC}}$ holds with very good accuracy, Also the obtained value of the scaling coefficient 0.9 $\mathring{A}^{-2}$ is close to the experimental value $\epsilon_{\mathrm{SM}}/r_c^2\approx 2$ $\mathring{A}^{-2}$ (for $\epsilon_{\mathrm{SM}}\approx 80$ and $r_{c_{\mathrm{SM}}}\approx 1.7a$). That is, the estimated values of R_c^2 approximately two times larger than the corresponding experimental values shown in Fig. 4 (e.g., at $T=300$ K $R_c^{\mathrm{theor}}\sim 190$ Å compared with $R_c^{\mathrm{exper}}\sim 130$ Å).

Thus we conclude that the size of PNR in relaxor ferroelectrics could be estimated from the rather universal interrelation (23) between the parameters of the soft mode dispersion curve and the static dielectric permittivity. In Ref. 50 the proportionality $I(q=0)\propto R_c^2$ of the neutron diffuse scattering intensity has been observed in agreement with Eqs. (22) and (23) and contrary to the authors' expectations $I(q=0)\propto S_{q=0}\propto TR_c^2$ based on the improper use of the fluctuation-dissipation theorem.[50,51]

In fact, according to the fluctuation-dissipation theorem we have in the mean field regime

$$S_{q=0} = \frac{T(\epsilon_{\mathrm{FC}}-\epsilon_l)}{4\pi} = \frac{T_c\epsilon_l}{4\pi\left(1-\dfrac{T_c}{T}\right)} = \frac{nd^{*2}}{3\epsilon_l}\epsilon_{\mathrm{FC}}, \tag{50}$$

which coincides with Eq. (22) at $q=0$, and results in $I(q=0)\propto R_c^2$.

Also, as we mentioned in Sec. III B, the effect of quenched random fields on the structure factor cannot be treated with use of the fluctuation-dissipation theorem, and instead the approach developed in Sec. III B should be used.

C. Interrelation between NMR spin lattice relaxation and frequency dependent dielectric response

As mentioned in Sec. IV C, NMR spin lattice relaxation rate T_1^{-1} in ferroelectrics is proportional to the autocorrelation

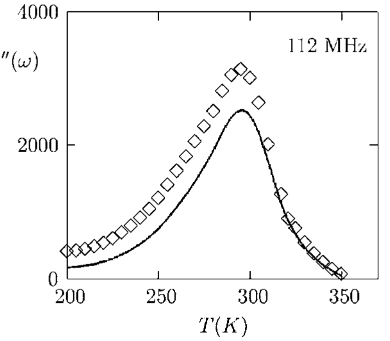

FIG. 5. Experimental (squares) and theoretical (solid line) temperature dependence of the imaginary part of dielectric permittivity in PMN. The solid line is calculated from Eq. (48) using the experimental data (Ref. 26) on the real part of dielectric permittivity and the values of Q''_ω presented in Fig. 6.

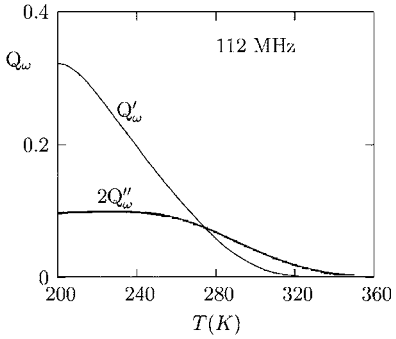

FIG. 6. The values of the real and imaginary part of the spectral function Q_ω. The values of Q'_ω have been reconstructed from Eq. (47) using the experimental data of ϵ' (Ref. 26). The values of Q''_ω correspond to the best fit of the theory with both experiments presented in Figs. 1 and 5.

function of polarization $G^{\text{loc}}_{\Omega_{\text{Larm}}}$. Thus, the interrelation between T_1 and $\epsilon''(\Omega_{\text{Larm}})$ provides a meaningful test to probe the different models of relaxor dynamics, since these quantities depend on different parts of polar fluctuation spectrum.

The results of the theory for the temperature dependence of T_1 are presented in Fig. 1 along with the experimental results[20] on T_1 and ϵ''. One can see from Fig. 1 that T_1 deviates significantly from ϵ'', which shows that dynamics of local and long wave length polar fluctuations are quite different. In particular, the time scale of the local polarization dynamics at low temperatures is shorter than the time scale of the average polarization dynamics of PNR. The same tendency has been observed in Ref. 21 for ^{139}La NMR spin lattice relaxation in PLZT (however, the authors[21] ascribed the difference between ϵ'' and T_1 to the contribution of nonpolar atomic motion rather than to the diffusion type propagation of the local polarization).

The solid line in Fig. 1 has been calculated from the continuous time random walk model, Eq. (44), which takes into account diffusion type polarization propagation inside each PNR. Since the probability density $g(\tau_i)$ of cluster relaxation times is unknown, one could consider the function $Q''_\omega(T)$ in Eq. (44) as a fitting function which determines both imaginary part of permittivity ϵ'' shown in Fig. 5, and NMR spin lattice relaxation time T_1.

The function $Q''_\omega(T)$ used in the fit is presented in Fig. 6, along with the function $Q'_\omega(T)$, which has been reconstructed from Eq. (47) using experimental data[26] on the real part of permittivity at frequency $\omega = 112$ MHz, which is close to ^{207}Pb Larmor frequency 79.4 MHz. The data for $\epsilon_{\text{FC}}(T)$ has been obtained from Ref. 26 using the values of ϵ' for lower frequencies and interpolation procedure similar to that discussed in Ref. 15. The value $\epsilon_l = 1000$ has been used according to the discussion in Sec. IV A.

It is apparent that due to approximations involved (especially, neglecting the correlations between relaxation times of different polar clusters) one could not expect that the same function $Q''_\omega(T)$ would reproduce the exact temperature dependence of both ϵ'' and T_1. However, as one can see from Figs. 1 and 5, there is a reasonable agreement between the theory and the experiment, which strongly supports the proposed model of relaxor behavior of PMN.

At the same time the deviation of the theory from the experiment indicates that the correlation effects between relaxation times of different polar clusters are not negligible, and should be taken into account for the deeper understanding of the complex relaxor dynamics.

Note also that the technique adopted in Refs. 33 and 26 for solving inverse problem on reconstructing the probability density of relaxors from experimental data could be applied for the reconstruction of cluster relaxation time probability density $g(\tau)$ from the values $Q'_\omega(T)$, $Q''_\omega(T)$. The latter could be extracted from dielectric measurements at different frequencies.

VI. CONCLUSION

We have shown that the dynamic behavior of PNR in relaxor ferroelectrics could be understood within the model of interacting short range polar clusters induced by off-center ions in highly polarizable materials. The polarization dynamics is controlled by two main factors: (a) distribution of cluster reorientation frequencies contributing to the local glass like freezing and (b) distribution of the local fields originated from quenched random field fluctuations controlling the effect of the critical slowing down.

The theory reveals the origin of PNR and gives a simple estimate for the PNR size from a relationship involving the parameters of the soft mode dispersion curve and static dielectric permittivity. The neglect of correlations between relaxation times of different short range clusters incorporated with the continuous time random walk framework allowed to obtain the dynamic equations for local polarization dynamics and establish the relationship between NMR spin lattice relaxation rates and frequency dependent dielectric response.

From the comparison between the theory and the experiment in PMN we conclude that the polar clusters within PNR are not tight binding entities, and diffusion type propagation modes control their local polarization dynamics. At the same time, some deviation of the theory from the experiment indicates that the correlation effects between relaxation times of different polar clusters are not negligible, and should be taken into account for the deeper understanding of the complex relaxor dynamics.

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