

RELAXATION DYNAMICS OF DISORDERED ISING MODEL. TWO-SITE CLUSTER APPROXIMATION

R.R.LEVITSKII, S.I.SOROKOV, R.O.SOKOLOVSKII

*Institute for Condensed Matter Physics
Ukrainian National Academy of Sciences
1 Svientsitski St., 290011 Lviv-11, Ukraine*

Received September 12, 1995

Spin relaxation in a site-disordered Ising model within master equation approach is studied. The \vec{q}, ω -dependent susceptibility of the model is calculated and investigated. Effects described by the two-site cluster approximation and lost by the mean field approximation are discussed. Comparison of obtained results to dielectric measurements in $Cs(H_{1-x}D_x)_2PO_4$ is presented.

1. Introduction

Being the model of critical phenomena in many different objects an Ising model compels an attention during many years. This paper investigate the kinetics of the model (within master equation [1] approach) having several goals. We consider the disordered (site chaos) Ising model in order to describe the dynamic properties ferroelectrics of partially deuterated with hydrogen bonds. Besides the theory seems to be applicable to many other systems: alloys of magnets, diluted magnetic or ferroelectric systems and others. Within two-site cluster approximation (TCA) we calculate the dynamic structure factor to supplement our previous study [2] of correlation functions for the model.

2. The Hamiltonian. General approach

We consider the spin system with site chaos on the Bravais lattice which is described by the Ising Hamiltonian

$$H = - \sum_i \kappa_i S_i - \frac{1}{2} \sum_{ij} K_{ij} S_i S_j . \quad (2.1)$$

H describes the set of Ising spins with the pair exchange interaction K_{ij} in the site-dependent field κ_i . The set of variables $\{S_i\}$ ($S_i = \pm 1$; $i = 1 \cdots N$, N is a number of the sites) represents a state of spin subsystem, and the set $\{X_{i\alpha}\}$ describes the sort configuration ($X_{i\alpha} = 1$, if the site i is occupied

by spin of the sort α , otherwise $X_{i\alpha} = 0$; $\alpha = 1 \cdots \Omega$, Ω is a number of the sorts). The quantities κ_i , K_{ij} depend on sort configuration

$$\kappa_i = \sum_{\alpha} \kappa_{i\alpha} X_{i\alpha}; \quad K_{ij} = \sum_{\alpha\beta} K_{i\alpha,j\beta} X_{i\alpha} X_{j\beta}, \quad (2.2)$$

so the true parameters of the Hamiltonian (2.1) are $\kappa_{i\alpha}$, $K_{i\alpha,j\beta}$. Here we suppose the pair interactions to be short-range

$$K_{i\alpha,j\beta} = K_{\alpha\beta} \pi_{ij}; \quad K_{\alpha\beta} > 0; \quad \pi_{ij} = \begin{cases} 1, & \text{if } j \in \pi_i, \\ 0, & \text{otherwise,} \end{cases} \quad (2.3)$$

where π_i denotes the set of the nearest neighbours of the site i (the first coordination sphere).

From the very beginning we shall consider the quenched system (frozen sort configuration) and investigate its dynamics on the basis of Glauber's master equation [1] for the distribution function $\rho(\{S\}, t)$:

$$\begin{aligned} \frac{d}{dt} \rho(\{S\}, t) &= - \sum_i W_i(\cdots S_i \cdots) \rho(\cdots S_i \cdots, t) \\ &+ \sum_i W_i(\cdots - S_i \cdots) \rho(\cdots - S_i \cdots, t). \end{aligned} \quad (2.4)$$

Following ref. [1] the probability per unit time that the i th spin flips from the value S_i to $-S_i$ is assumed to be

$$W_i(\cdots S_i \cdots) = \frac{1}{2\tau_i} (1 - S_i \tanh \beta \varepsilon_i), \quad (2.5)$$

where

$$\varepsilon_i = \kappa_i + \sum_j K_{ij} S_j \quad (2.6)$$

is a local effective field acting on the i th spin. In this approach the dissipative properties of the spin subsystem is caused by its interaction with another (e.g. phonon) subsystems which play the role of a thermal bath. The quantity τ_i has the dimension of time and defines the time scale of relaxational phenomena. We assume the quantity τ_i to be locally sort-dependent:

$$\tau_i = \sum_{\alpha} \tau_{\alpha}^0 X_{i\alpha}. \quad (2.7)$$

Relations (2.4) and (2.5), (2.6) lead to the following kinetic equations for the average value of spin:

$$D_{i,t} \langle S_i \rangle_{H,t} = \langle \tanh \beta \varepsilon_i \rangle_{H,t}, \quad (2.8)$$

where

$$\langle (\cdots) \rangle_{H,t} = S p_s \rho(\{S\}, t) (\cdots); \quad D_{i,t} = 1 + \tau_i \frac{d}{dt}. \quad (2.9)$$

The system is disordered, therefore in order to calculate experimentally measured quantities one have to perform averaging over the sort configurations. For example, the polarization of a partially deuterated H-bonded ferroelectric with the Hamiltonian (2.1) is defined as

$$P(t) = \frac{1}{V} \sum_i \sum_{\alpha} \mu_{\alpha} \langle \langle S_{i\alpha} \rangle_{H,t} \rangle_x; \quad S_{i\alpha} = S_i X_{i\alpha}, \quad (2.10)$$

where V is a volume of the system, μ_α the dipole electric moment associated with a quasispin of sort α (proton or deuteron), $\langle \cdots \rangle_x$ denotes averaging over sort configurations with some density matrix $\rho(\{X_{i\alpha}\})$ that depends on conditions of system's freezing. The two-site cluster approximation, which is used below, is sensitive only to the following moments of the distribution $\rho(\{X_{i\alpha}\})$

$$\langle X_{i\alpha} \rangle_x = c_\alpha, \quad \langle X_{i\alpha} X_{j\beta} \rangle_x = w_{\alpha\beta} \quad (j \in \pi_i), \quad (2.11)$$

where c_α has the meaning of the sort α spins' concentration.

3. Formulation of the two-site cluster approximation

The equation (2.8) is quite intractable. One can expand the $\langle \tanh \beta \varepsilon_i \rangle_{H,t}$ in the right-hand side of (2.8) and find that this equation couples correlation functions (CFs) of the type $\langle S_{i_1} \cdots S_{i_n} \rangle_{H,t}$ where the site indices belong to the first coordination sphere of the site i ($i_1 \cdots i_n \in \pi_i$). The equation for $\langle S_{i_1} \cdots S_{i_n} \rangle_{H,t}$ derived from (2.4) and (2.5) involves higher CFs, so the exact treatment of the Glauber's equations has succeeded only in the case of one-dimensional system in zero field (ideal (one-sort) chain [1,3,4], chain with single impurity [5]). In other cases interpolative approximations are used which express higher CFs via lower, e.g. $\langle \prod_j S_j \rangle_{H,t} \approx \prod_j \langle S_j \rangle_{H,t}$.

We suggest here the closure to the equation (2.8) in the spirit of a cluster approach. In a one-site approximation we replace contributions of all spins with effective fields

$$\varepsilon_i \rightarrow \varepsilon_i^{[1]} = \bar{\kappa}_{i,t} + \sum_{r \in \pi_i} r \bar{\varphi}_{i,t} \equiv \bar{\kappa}_{i,t}, \quad (3.1)$$

where ${}_r \bar{\varphi}_{i\alpha}$ has the meaning of effective field acting on the spin i of sort α from the nearest neighbour on the site r . In a two-site approximation a contribution of one of the nearest neighbours is taken into account explicitly

$$\varepsilon_i \rightarrow \varepsilon_i^{[2]} = \bar{\kappa}_{i,t} + \sum_{\substack{r \in \pi_i \\ r \neq 2}} {}_r \bar{\varphi}_{i,t} + K_{ij} S_j \equiv {}_j \bar{\kappa}_{i,t} + K_{ij} S_j, \quad (j \in \pi_i). \quad (3.2)$$

In the case of ideal (one-sort) system no more assumptions are needed to obtain the closed set of equations for the average value of spin $\langle S_i \rangle_{H,t}$

$$\begin{aligned} D_{i,t} \langle S_i \rangle_{H,t} &= \langle \tanh \beta \varepsilon_{i,t}^{[1]} \rangle_{H,t}, \\ D_{i,t} \langle S_i \rangle_{H,t} &= \langle \tanh \beta \varepsilon_{i,t}^{[2]} \rangle_{H,t}. \end{aligned} \quad (3.3)$$

For the many-sort system it is necessary to point out a dependence on the fields ${}_r \bar{\varphi}_{i,t}$ on the sort configuration. We shall believe the fields ${}_r \bar{\varphi}_{i,t}$ to be different in the one-site approximations and the two-site one until averaging over sort configurations is carried out. For the sort averaging procedure both "one-site" and "two-site" fields will be assumed to be locally sort-dependent

$${}_r \bar{\varphi}_{i,t} = \sum_{\alpha} X_{i\alpha} {}_r \bar{\varphi}_{i\alpha,t}, \quad (3.4)$$

where ${}_r \bar{\varphi}_{i\alpha,t}$ is a c-number (it does not contain spin or sort operators).

After simple transformation the equation (3.3) can be written in the form

$$\begin{aligned} D_{i,t}\langle S_i \rangle_{H,t} &= \tanh \beta \bar{\kappa}_{i,t}, \\ D_{i,t}\langle S_i \rangle_{H,t} &= L_{ij,t} + P_{ij,t}\langle S_j \rangle_{H,t}, \\ D_{j,t}\langle S_j \rangle_{H,t} &= L_{ji,t} + P_{ji,t}\langle S_i \rangle_{H,t}; \quad (j \in \pi_i) \end{aligned} \quad (3.5)$$

where

$$\begin{aligned} L_{ij,t} &= \frac{1}{2} [\tanh \beta({}_j\bar{\kappa}_{it} + K_{ij}) + \tanh \beta({}_j\bar{\kappa}_{it} - K_{ij})], \\ P_{ij,t} &= \frac{1}{2} [\tanh \beta({}_j\bar{\kappa}_{it} + K_{ij}) - \tanh \beta({}_j\bar{\kappa}_{it} - K_{ij})]. \end{aligned} \quad (3.6)$$

The expressions (3.5) compose a system of nonlinear differential equations. To linearize these expressions we consider only linear response of the system to a small time-dependent change of the external field. For this purpose we write the dynamic quantities in the following form:

$$\begin{aligned} \kappa_{i,t} &= \kappa_i + \delta\kappa_{i,t}; \quad \bar{\kappa}_{i,t} = \bar{\kappa}_i + \delta\bar{\kappa}_{i,t}; \\ {}_j\bar{\kappa}_{i,t} &= {}_j\bar{\kappa}_i + \delta{}_j\bar{\kappa}_{i,t}; \quad \langle S_i \rangle_{H,t} = m_i^{(1)} + \delta m_{i,t}^{(1)}; \\ L_{ij,t} &= L_{ij} + L'_{ij} \delta{}_j\bar{\kappa}_{i,t}; \quad P_{ij,t} = P_{ij} + P'_{ij} \delta{}_j\bar{\kappa}_{i,t}; \\ \tanh \beta \bar{\kappa}_{i,t} &= \tanh \bar{\kappa}_i + (1 - \tanh^2 \bar{\kappa}_i) \delta\bar{\kappa}_{i,t}. \end{aligned} \quad (3.7)$$

Here κ_i , $\bar{\kappa}_i$, ${}_j\bar{\kappa}_i$, $m_i^{(1)}$, L_{ij} , P_{ij} are the static (independent of time) components of corresponding quantities and $\delta\bar{\kappa}_{i,t}$, $\delta{}_j\bar{\kappa}_{i,t}$, $\delta m_{i,t}^{(1)}$, $L'_{ij} \delta{}_j\bar{\kappa}_{i,t}$, $P'_{ij} \delta{}_j\bar{\kappa}_{i,t}$, represent linear in $\delta\kappa_{i,t}$ deviations of these quantities from their static values.

$$\begin{aligned} L_{ij} &= \frac{1}{2} (\tanh \beta({}_j\bar{\kappa}_i + K_{ij}) + \tanh \beta({}_j\bar{\kappa}_i - K_{ij})), \\ P_{ij} &= \frac{1}{2} (\tanh \beta({}_j\bar{\kappa}_i + K_{ij}) - \tanh \beta({}_j\bar{\kappa}_i - K_{ij})), \\ L'_{ij} &= \frac{1}{2} (\cosh^{-2} \beta({}_j\bar{\kappa}_i + K_{ij}) + \cosh^{-2} \beta({}_j\bar{\kappa}_i - K_{ij})), \\ P'_{ij} &= \frac{1}{2} (\cosh^{-2} \beta({}_j\bar{\kappa}_i + K_{ij}) - \cosh^{-2} \beta({}_j\bar{\kappa}_i - K_{ij})). \end{aligned} \quad (3.8)$$

Due to (3.7) two independent systems of equations are obtained from (3.5). The static part:

$$\begin{aligned} m_i^{(1)} &= \tanh \beta \bar{\kappa}_i, \\ m_i^{(1)} &= L_{ij} + P_{ij} m_j^{(1)}; \quad m_j^{(1)} = L_{ji} + P_{ji} m_i^{(1)} \end{aligned} \quad (3.9)$$

can be reduced to the form

$$m_i^{(1)} = \tanh \beta \bar{\kappa}_i = \frac{L_{ij} + P_{ij} L_{ji}}{1 - P_{ij} P_{ji}}. \quad (3.10)$$

The same equation is also obtained on the basis of cluster expansion of the free energy [6] and can be written in the form

$$m_i^{(1)} = \langle S_i \rangle_{\rho_i} = \langle S_i \rangle_{\rho_{ij}}, \quad (3.11)$$

where the one-site ρ_i and intracluster ρ_{ij} density matrices are defined as follows

$$\begin{aligned}\rho_i &= \frac{\exp(-\beta H_i)}{Sp_{S_i} \exp(-\beta H_i)}; H_i = \bar{\kappa}_i S_i, \\ \rho_{ij} &= \frac{\exp(-\beta H_{ij})}{Sp_{S_i S_j} \exp(-\beta H_{ij})}; H_{ij} = {}_j\bar{\kappa}_i S_i + {}_i\bar{\kappa}_j S_j + K_{ij} S_i S_j.\end{aligned}\quad (3.12)$$

Multiplying (3.10) by $X_{i\alpha}$ and averaging the obtained relation over sort configurations we get the equation for an average value of sort α spins $m_{i\alpha}^{(1)} \equiv \langle m_i^{(1)} X_{i\alpha} \rangle_x$

$$\begin{aligned}m_{i\alpha}^{(1)} &= c_\alpha \tanh \bar{\kappa}_{i\alpha}, \\ &= \sum_\beta w_{\alpha\beta} \frac{\sinh \beta({}_j\bar{\kappa}_{i\alpha} + {}_i\bar{\kappa}_{j\beta}) + a_{\alpha\beta} \sinh \beta({}_j\bar{\kappa}_{i\alpha} - {}_i\bar{\kappa}_{j\beta})}{\cosh \beta({}_j\bar{\kappa}_{i\alpha} + {}_i\bar{\kappa}_{j\beta}) + a_{\alpha\beta} \cosh \beta({}_j\bar{\kappa}_{i\alpha} - {}_i\bar{\kappa}_{j\beta})}.\end{aligned}\quad (3.13)$$

On assuming uniform field $\kappa_{i\alpha} \rightarrow \kappa_\alpha$ (UF: ${}_r\bar{\varphi}_{i\alpha} \rightarrow \bar{\varphi}_\alpha$, ${}_j\bar{\kappa}_{i\alpha} \rightarrow \kappa_\alpha + (z-1)\bar{\varphi}_\alpha \equiv \bar{\kappa}'_\alpha$, $\bar{\kappa}_{i\alpha} \rightarrow \kappa_\alpha + z\bar{\varphi}_\alpha \equiv \bar{\kappa}_\alpha$, z is a first coordination number) we obtain the known in corresponding static theories (TCA [2], first order of cluster variation method [7]) relation: (3.13) becomes the set of 2Ω equations for the same number of quantities – Ω order parameters $m_\alpha^{(1)}$ and Ω effective fields $\bar{\varphi}_\alpha$.

The dynamic linearized part of (3.5) (here nonlinear on $\delta\kappa_{i,t}$ terms $L'_{ij} \delta m_{i,t}^{(1)} \delta {}_i\bar{\kappa}_{j,t}$, $L'_{ji} \delta m_{j,t}^{(1)} \delta {}_j\bar{\kappa}_{i,t}$ should be neglected) after time Fourier transformation and averaging over sort configurations yields the Ornstein-Zernike-type equation for the dynamic structure factor $m_{i\alpha,j\beta}^{(2)}(\omega) = \frac{\delta m_{i\alpha}^{(1)}(\omega)}{\delta \kappa_{j\beta}(\omega)}$ of the model. Spatial Fourier transformation presents its solution in the form

$$\begin{aligned}(\hat{m}^{(2)}(\vec{q}, \omega))^{-1} &= (1-z) (\hat{\phi}^{(2)})^{-1} + z (\hat{\phi}^{(20)} + \hat{\phi}^{(11)})^{-1} \\ &+ (\pi_{\vec{0}} - \pi_{\vec{q}}) \left(\hat{\phi}^{(20)} (\hat{\phi}^{(11)})^{-1} \hat{\phi}^{(20)} - \hat{\phi}^{(11)} \right)^{-1},\end{aligned}\quad (3.14)$$

where matrix elements are

$$\begin{aligned}(\hat{\phi}^{(2)})_{\alpha\beta} &\stackrel{UF}{=} \delta_{\alpha\beta} c_\alpha \frac{1 - \tanh^2 \beta \bar{\kappa}_\alpha}{D_\alpha}, \\ (\hat{\phi}^{(20)})_{\alpha\beta} &\stackrel{UF}{=} \delta_{\alpha\beta} \sum_\gamma w_{\alpha\gamma} \frac{R_{\alpha\gamma} D_\gamma}{D_\alpha D_\gamma - P_{\alpha\gamma} P_{\gamma\alpha}}, \\ (\hat{\phi}^{(11)})_{\alpha\beta} &\stackrel{UF}{=} w_{\alpha\beta} \frac{R_{\beta\alpha} P_{\alpha\beta}}{D_\alpha D_\beta - P_{\alpha\beta} P_{\beta\alpha}}, \\ (\hat{m}^{(2)}(\vec{q}, \omega))_{\alpha\beta} &= \sum_j e^{i\vec{q}(\vec{R}_j - \vec{R}_i)} m_{i\alpha,j\beta}^{(2)}(\omega),\end{aligned}\quad (3.15)$$

and

$$P_{\alpha\beta} = \frac{1}{2} (\tanh \beta(\bar{\kappa}'_\alpha + K_{\alpha\beta}) - \tanh \beta(\bar{\kappa}'_\alpha - K_{\alpha\beta})), \quad (3.16)$$

$$\begin{aligned}
R_{\alpha\beta} = & 2a_{\alpha\beta} \left[2a_{\alpha\beta} (1 + \cosh 2\beta\bar{\kappa}'_{\alpha} \cosh 2\beta\bar{\kappa}'_{\beta}) + \right. \\
& \left. (1 + a_{\alpha\beta}^2) (\cosh 2\beta\bar{\kappa}'_{\alpha} + \cosh 2\beta\bar{\kappa}'_{\beta}) \right] \times \\
& \left[(1 + a_{\alpha\beta}^2 + 2a_{\alpha\beta} \cosh 2\beta\bar{\kappa}'_{\alpha}) \right]^{-1} \times \\
& \left[\cosh \beta(\bar{\kappa}'_{\alpha} + \bar{\kappa}'_{\beta}) + a_{\alpha\beta} \cosh \beta(\bar{\kappa}'_{\alpha} - \bar{\kappa}'_{\beta}) \right]^{-2},
\end{aligned}$$

Factor $\pi_{\vec{q}}$ has the form

$$\pi_{\vec{q}} = \sum_{j \in \pi_i} e^{i\vec{q}(\vec{R}_j - \vec{R}_i)}; \quad \pi_{\vec{0}} = z. \quad (3.17)$$

4. Annealed model

In this case protons (deuterons) can move from one site (hydrogen bond) to another. Therefore the sort configuration changes in time. This motion should be described by Kawasaki master equation [9]. But we assume that the sort configuration changes slowly and for the frequencies of magnetic (or ferroelectric) dispersion, quasispins see the sort configuration at rest. Therefore the formula (3.14) remains valid. If the temperature changes quite slowly, $w_{\alpha\beta}$ is a temperature-dependent quantity which can be found by the static theory [2].

5. Discussion. Numerical results.

The expression (3.14) is our target formula. Let us consider its partial cases. All expressions will be given for UF case.

1) *Noninteracting system* ($K_{\alpha\beta} = 0 \forall \alpha, \beta$) is a trivial case.

$$m_{\alpha}^{(1)} = c_{\alpha} \tanh \beta\kappa_{\alpha}, \quad (5.1)$$

$$m_{\alpha\beta}^{(2)}(\vec{q}, \omega) = \delta_{\alpha\beta} \frac{c_{\alpha}(1 - \tanh^2 \beta\kappa_{\alpha})}{1 + i\omega\tau_{\alpha}^0}. \quad (5.2)$$

All the spins are independent, so the structure factor are independent of the wavelength. The relaxation times are equal to τ_{α}^0 .

2) *One-sort (ideal) system*. This case can be obtained in the limit $c_1 = 1$. The single order parameter $m_1^{(1)}$ and the effective field φ_1 satisfy the following equation

$$m_1^{(1)}/c_1 \equiv \sigma_1^{(1)} = \tanh \beta\bar{\kappa}_1 = \frac{\sinh 2\beta\bar{\kappa}'_1}{\cosh 2\beta\bar{\kappa}'_1 + a_{11}}, \quad (5.3)$$

where

$$\bar{\kappa}_1 = \kappa_1 + z\bar{\varphi}_1; \quad \bar{\kappa}'_1 = \kappa_1 + z'\bar{\varphi}_1; \quad z' = z - 1; \quad a = e^{-2K}. \quad (5.4)$$

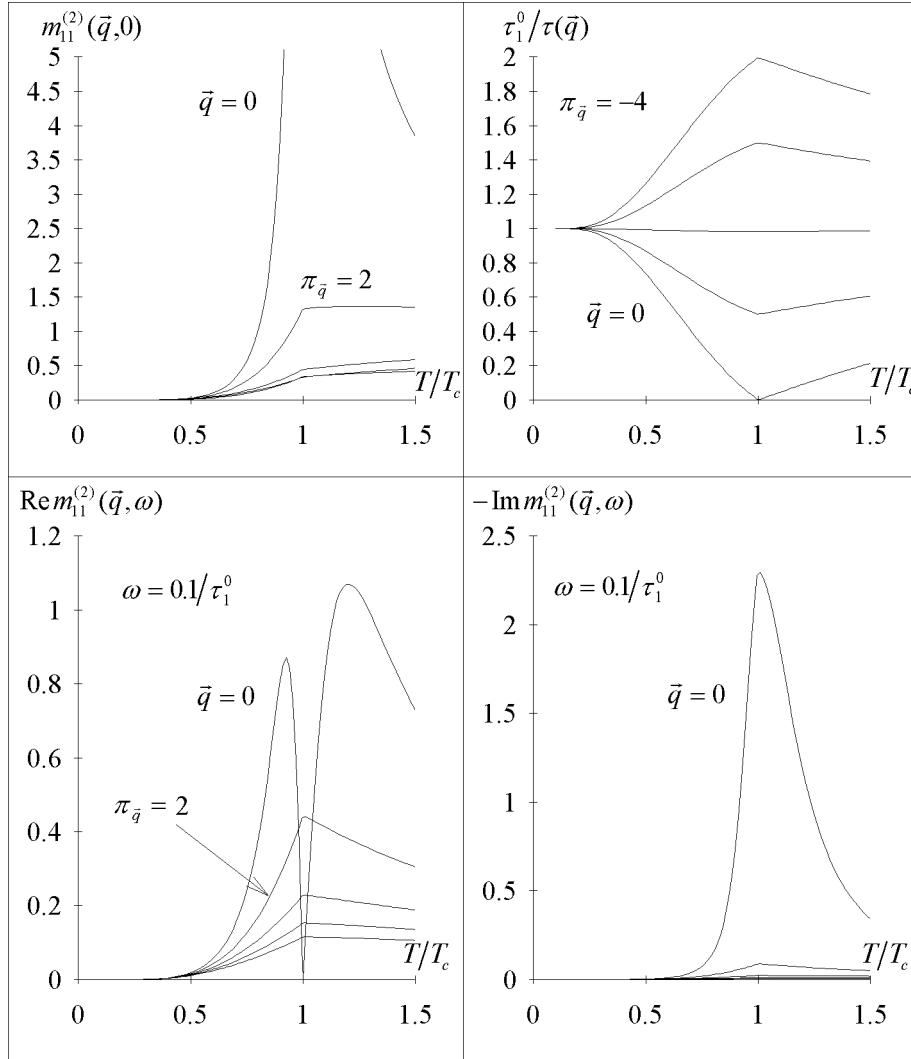


Figure 1: Pure system on plane square lattice ($z = 4$). Static pair CF $m_{11}^{(2)}(\vec{q}, 0)$, inverse relaxation time $1/\tau(\vec{q})$, real and imaginary parts of dynamic pair CF $m_{11}^{(2)}(\vec{q}, \omega)$ at different wavevectors: $\pi_{\vec{q}}$ equals 4 ($\vec{q} = 0$), 2, 0, -2, -4 (the last corresponds to the vertices of the first Brillouin zone).

The dynamic structure factor $m_{11}^{(2)}(\vec{q}, \omega)$ takes the form

$$m_{11}^{(2)}(\vec{q}, \omega)/c_1 \equiv \sigma_{11}^{(2)}(\vec{q}, \omega) = \frac{\sigma_{11}^{(2)}(\vec{q}, 0)}{1 + i\omega\tau(\vec{q})}, \quad (5.5)$$

where

$$\sigma_{11}^{(2)}(\vec{q}, 0) = \left(1 - (\sigma_1^{(1)})^2\right) \frac{1 - P^2}{1 + z'P^2 - P\pi_{\vec{q}}}, \quad (5.6)$$

$$P = \frac{1 - a^2}{1 + a^2 + 2a \cosh 2\beta\bar{\kappa}'} \frac{1}{1 - (\sigma_1^{(1)})^2},$$

and $\sigma_{11}^{(2)}(\vec{q}, 0)$ coincides with a spatial Fourier-transform of the static correlation function $\langle S_i S_j \rangle_H^c$, which was founded with cluster expansion of the free energy within TCA in the ref. [6]. Result (5.6) is accurate in the one-dimensional case. Relaxation time

$$\tau(\vec{q}) = \tau_1^0 \left(1 - \pi_{\vec{q}} \frac{P}{1 + z'P^2} \right)^{-1} \xrightarrow{T > T_c} \tau^0 \left(1 - \pi_{\vec{q}} \frac{\tanh \beta K}{1 + z' \tanh^2 \beta K} \right)^{-1} \quad (5.7)$$

is a decreasing function of \vec{q} . Note, that known exact result [3] (zero external field, ideal one-dimensional system) is a partial case of the formula (5.5).

Figure 1 demonstrates the temperature and \vec{q} -dependence of the static structure factor $m_{11}^{(2)}(\vec{q}, 0)$ and the relaxation time $\tau(\vec{q})$ of the system on the plane square lattice ($z = 4$). Five lines represent five wavevectors: $\pi_{\vec{q}} = 4, 2, 0, -2, -4$. In this figure (as well as in the formula (5.7)) one can see an interesting symmetry of inverse relaxation time: $\tau_1^0/\tau(\vec{q})$ line at $\pi_{\vec{q}}$ is mirror reflection of the line at $-\pi_{\vec{q}}$. The figure 1 shows also a behaviour of real and imaginary parts of the dynamic structure factor (at the frequency $\omega = 0.1/\tau_1^0$).

3. *Diluted system* (interacting spins (sort 1) are diluted by the noninteracting (nonmagnetic, nonferroelectric) impurities: $K_{1\alpha} = 0$ for all $\alpha \neq 1$) is interesting due to percolation phenomena in it: concentration of sort 1 spins there is neither infinite cluster of interacting spins and ordered phase below a certain concentration of sort 1 spins. The mean field approximation (MFA) fails to describe percolation phenomena [7]. At zero external field within MFA, $\sigma_1^{(1)}$ and pair CF per interacting spin $\sigma_{11}^{(2)}$ depend on temperature, concentration and interaction strength only via T/T_c^{MFA} ($T_c^{MFA} = zc_1 K_{11}/k_b$): $\sigma_1^{(1)} = \sigma_1^{(1)}(T/T_c^{MFA})$, $\sigma_{11}^{(2)} = \sigma_{11}^{(2)}(\vec{q}, \omega, T/T_c^{MFA})$. Therefore MFA predicts that $\sigma_1^{(1)}$ reaches its saturation value ($\sigma_1^{(1)} = 1$ at $T = 0$) at any concentration of interacting spins, and $m_{11}^{(2)}(\vec{q}, \omega)$ shows a single relaxation time behaviour. In TCA $T_c = \frac{2K_{11}}{k_B \ln \frac{z'w_{11} + c_1}{z'w_{11} - c_1}}$,

$$m_{11}^{(1)} = c_1 \tanh \beta \bar{\kappa}_1 = w_{11} \frac{\sinh 2\beta \bar{\kappa}'_1}{\cosh 2\beta \bar{\kappa}'_1 + a_{11}} + (c_1 - w_{11}) \tanh \beta \bar{\kappa}'_1 \quad (5.8)$$

and this takes place for the case $w_{1\alpha} = 0$ only, otherwise at zero temperature $\sigma_1^{(1)} < 1$ (due to the presence of finite size clusters of interacting spins) and becomes zero if $c_1 \geq z'w_{11}$ (below percolation point). In the case of complete chaos ($w_{\alpha\beta} = c_\alpha c_\beta$) TCA predicts percolation concentration $c_p = (z - 1)^{-1}$. We note that the Effective Field Approximation of Kaneyoshi et.al. [8] (magnetization and static susceptibility) also reproduces this property of the diluted system.

Dynamic pair CF contains three terms

$$m_{11}^{(2)}(\vec{q}, \omega) = \frac{A_0}{1 + i\omega\tau_1^0} + \frac{A_+(\vec{q})}{1 + i\omega\tau_+(\vec{q})} + \frac{A_-(\vec{q})}{1 + i\omega\tau_-(\vec{q})}, \quad (5.9)$$

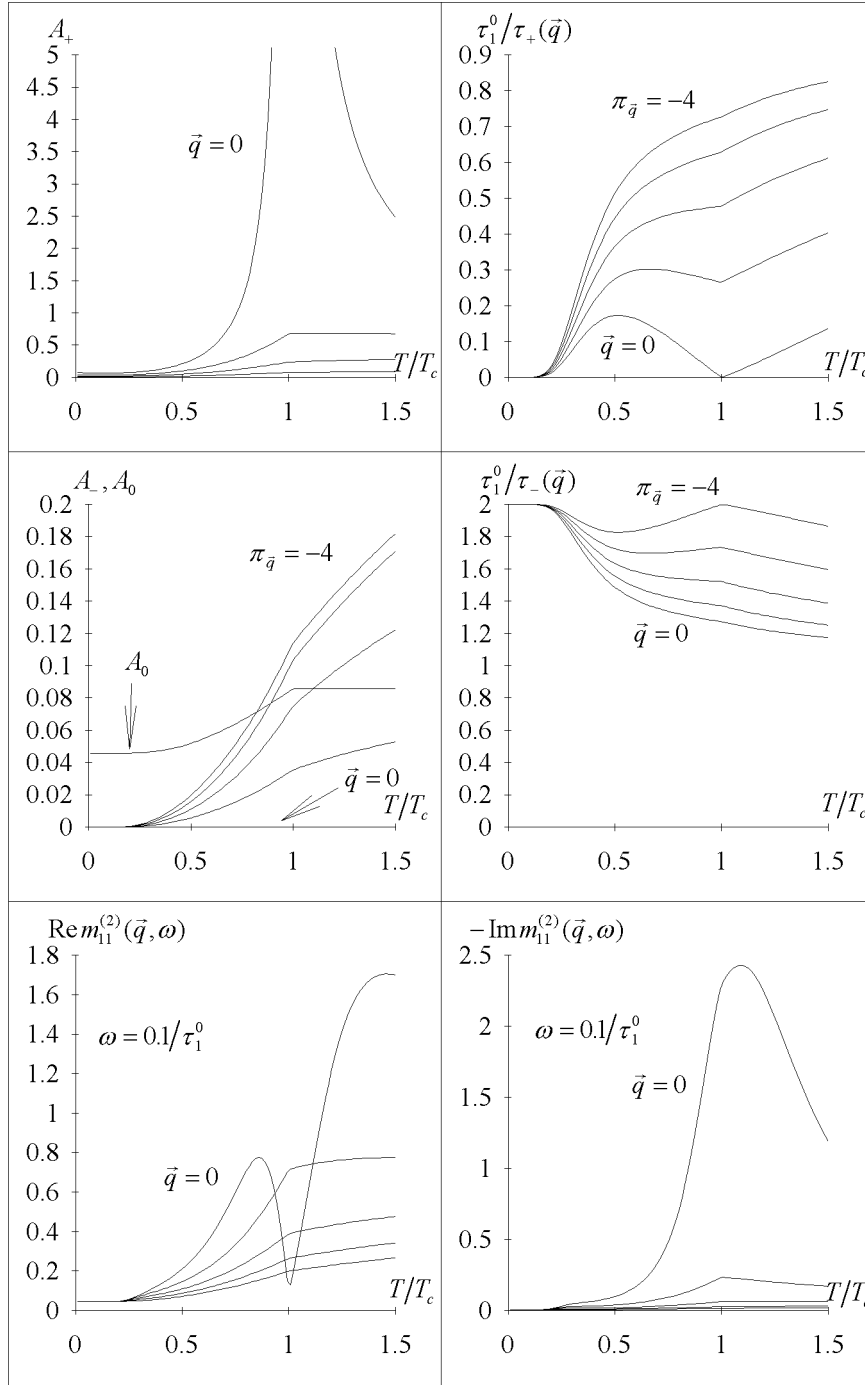


Figure 2: Diluted system ($c_1 = 0.6$, $w_{11} = c_1^2$) on plane square lattice (percolation concentration $c_p = 1/3$). Amplitudes A_+ , A_- , A_0 , inverse relaxation times $1/\tau_+(\vec{q})$, $1/\tau_-(\vec{q})$, and dynamic pair CF $m_{11}^{(2)}(\vec{q}, \omega)$ at different \vec{q} : $\pi_{\vec{q}} = 4, 2, 0, -2, -4$.

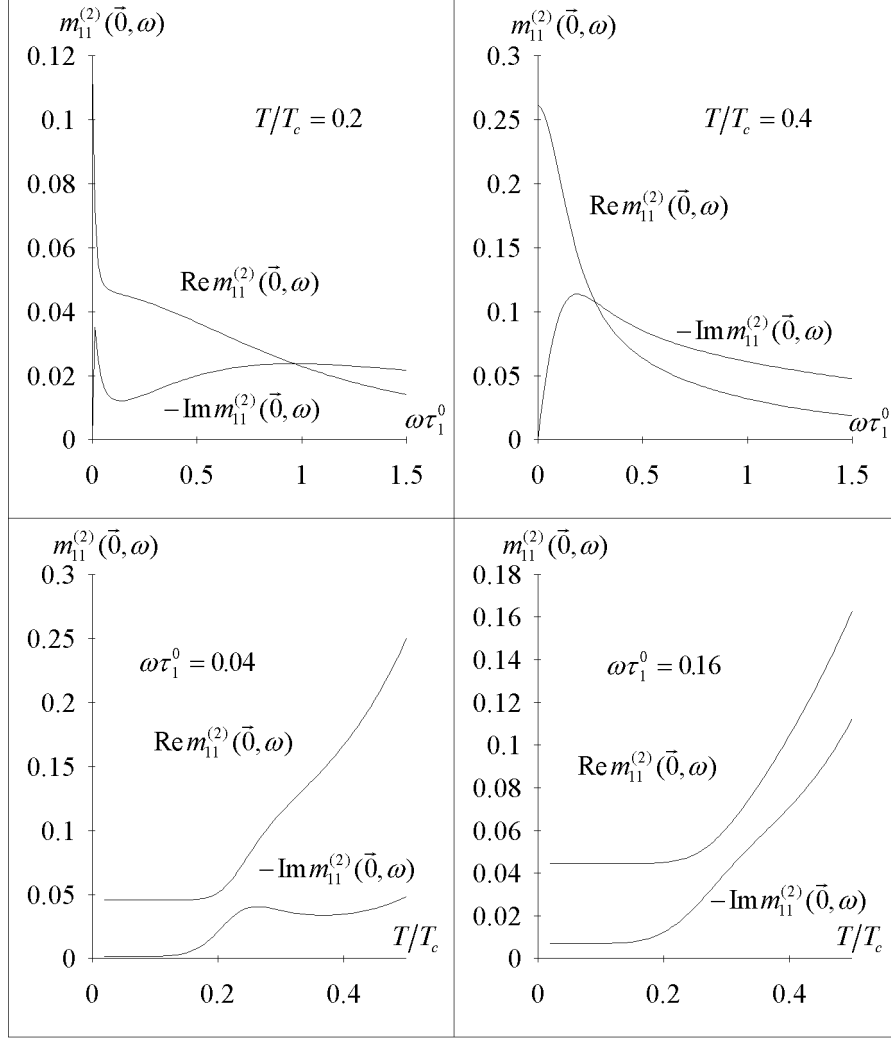


Figure 3: Diluted system. Temperature and frequency dependence of dynamic pair CF $m_{11}^{(2)}(\vec{q}, \omega)$ at $\vec{q} = 0$.

where

$$\begin{aligned}
 A_0 &= \frac{c_1 F_1^{(2)} w_{12} R_{12}}{z c_1 F_1^{(2)} - z' w_{12} R_{12}} \xrightarrow{T > T_c} \frac{c_1 w_{12}}{c_1 + z' w_{11}}, \\
 A_{\pm}(\pi_{\vec{q}}) &= \frac{B_{\pm}(\pi_{\vec{q}})}{1 - D_{\pm}(\pi_{\vec{q}})}; \quad \tau_{\pm}(\vec{q}) = \frac{\tau_1^0}{1 - D_{\pm}(\pi_{\vec{q}})}, \quad (5.10) \\
 B_{\pm}(\pi_{\vec{q}}) &= \pm \frac{c_1 F_1^{(2)}}{P D_{\pm}(\pi_{\vec{q}}) \sqrt{\mathcal{D}(\pi_{\vec{q}})}} (\langle R_1 \rangle^2 D_{\pm}^2(\pi_{\vec{q}}) - (P w_{12} R_{12})^2)
 \end{aligned}$$

and

$$\langle R_1 \rangle = w_{11} R_{11} + w_{12} R_{12},$$

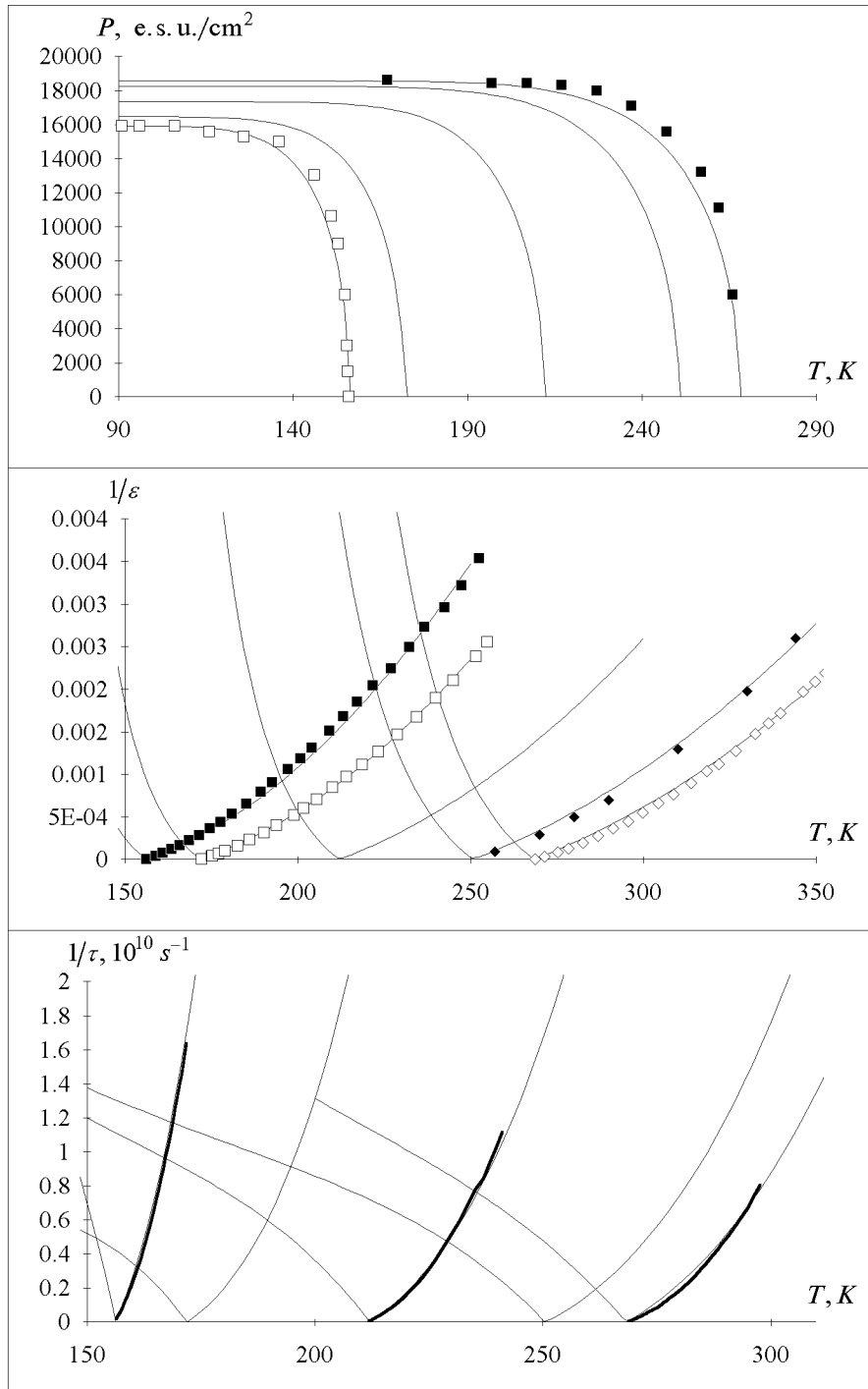


Figure 4: Spontaneous polarization P , inverse permittivity $1/\epsilon$ and inverse relaxation time $1/\tau$ of the ferroelectric $Cs(H_{1-x}D_x)_2PO_4$ for different degrees of deuteration. Squares correspond to experimental data of [12], bold lines to experimental points from the ref. [13]. Thick lines – theoretical results for annealed system with parameters given in table 1 and concentrations $x = 0, 0.18, 0.54, 0.88, 1$.

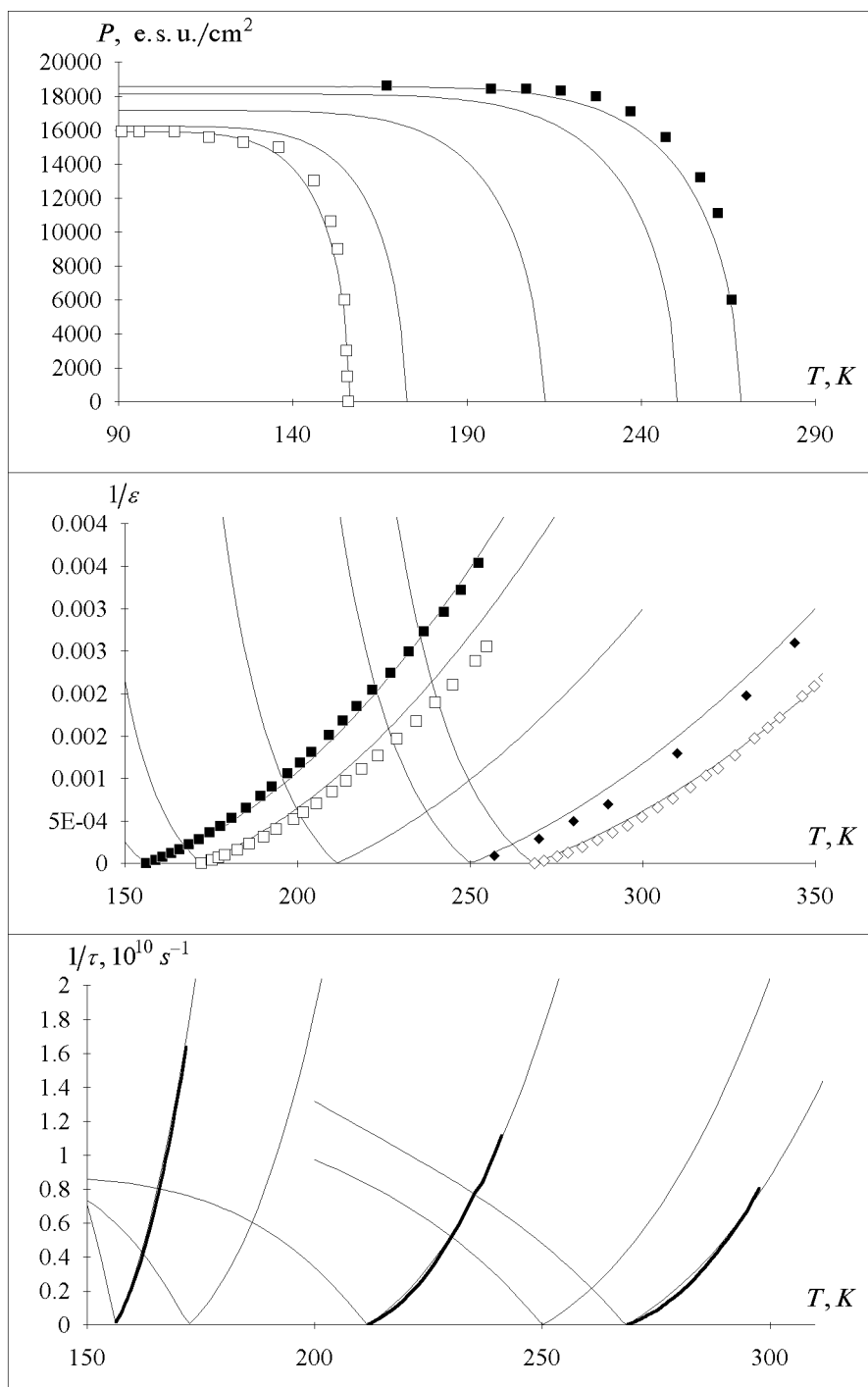


Figure 5: Comparison of experiments and quenched model with assumption $w_{\alpha\beta} = c_\alpha c_\beta$ (complete chaos) (see caption to the previous figure). Thick lines – theoretical results for quenched system with parameters given in table 1 and concentrations $x = 0, 0.12, 0.47, 0.83, 1$.

	quenched	annealed
$K_{11}/k_B, K$		610
$K_{12}/k_B, K$	500	500
$K_{22}/k_B, K$		390
$J_{11}/k_B, K$		2.85
$J_{12}/k_B, K$	3.2	1.6
$J_{22}/k_B, K$		1.06
$(V_{11} + V_{22} - 2V_{12})/k_B, K$	-	-1000
$I_{\alpha\beta}/k_B, K$	-	0
$\mu_1, e.s.u. \times cm$		2.18×10^{-18}
$\mu_2, e.s.u. \times cm$		1.87×10^{-18}
τ_1^0, s		3×10^{-14}
τ_2^0, s		5.5×10^{-15}

Table 1. The model parameters describing experimental data on the pseudo-one-dimensional ferroelectric $Cs(H_{1-x}D_x)_2PO_4$: short-range couplings $K_{\alpha\beta}$; long-range interactions $J_{\alpha\beta}$; effective dipole electric moments μ_α associated with proton or deuteron on a hydrogen bond; parameters of the master equation – relaxation times τ_α^0 of noninteracting protons or deuterons. $V_{\alpha\beta}$, $I_{\alpha\beta}$ are parameters describing nonexchange interactions between pseudospins [2]. Deuterons correspond to the pseudospins of sort 1.

$$\begin{aligned}
D_\pm(\pi_{\vec{q}}) &= P \frac{c_1 F_1^{(2)} w_{11} R_{11} \pm \sqrt{\mathcal{D}(\pi_{\vec{q}})}}{2\langle R_1 \rangle (z c_1 F_1^{(2)} - z' \langle R_1 \rangle)}, \\
\mathcal{D}(\pi_{\vec{q}}) &= (c_1 F_1^{(2)} w_{11} R_{11} \pi_{\vec{q}})^2 \\
&\quad + 4\langle R_1 \rangle (z c_1 F_1^{(2)} - z' \langle R_1 \rangle) w_{12} R_{12} (z c_1 F_1^{(2)} - z' R_{12}). \\
F_1^{(2)} &= c_1 (1 - (\sigma_1^{(1)})^2), \\
R_{12} &= 1 - \tanh^2 \beta \bar{\kappa}'_1, \\
R_{11} &= 4a \frac{1 + a \cosh 2\beta \bar{\kappa}'_1}{(\cosh 2\beta \bar{\kappa}'_1 + a)(1 + a^2 + 2a \cosh 2\beta \bar{\kappa}'_1)}, \\
P &= \frac{1}{2} [\tanh \beta(\bar{\kappa}'_1 + K) - \tanh \beta(\bar{\kappa}'_1 - K)]. \tag{5.11}
\end{aligned}$$

The first term of (5.9) has the (5.2)-like behaviour (relaxation time equals τ_1^0), so we believe it to be a contribution of isolated spins (i.e. the spins, which all neighbours are noninteracting). The last two terms of (5.9) have the following symmetry

$$\begin{aligned}
B_\pm(\pi_{\vec{q}}) &= B_\mp(-\pi_{\vec{q}}); \quad A_+(-z) = A_-(z) = 0, \\
D_\pm(\pi_{\vec{q}}) &= -D_\mp(-\pi_{\vec{q}}); \quad D_+(\pi_{\vec{q}}) > 0; \quad D_-(\pi_{\vec{q}}) < 0. \tag{5.12}
\end{aligned}$$

The relaxation time τ_- is always less than τ_1^0 while τ_+ is always greater than τ_1^0 and slows down at the critical point ($\tau_+(\vec{0}) \xrightarrow{T \rightarrow T_c} \infty$, $A_+(\pi_{\vec{0}}) \xrightarrow{T \rightarrow T_c} \infty$). Both τ_- and τ_+ increase when \vec{q} approaches to the center of the first Brillouin zone ($\vec{q} \rightarrow 0$). The amplitude A_- is equal to 0 in the centre of the first

Brillouin zone ($\pi_{\vec{q}} = z$) while A_+ vanishes at $\pi_{\vec{q}} = -z$ (in the vertices of the first Brillouin zone).

In the case $w_{11} = c_1$ ($w_{1\alpha} = 0 \forall \alpha \neq 1$, all interacting spins belong to one infinite cluster), A_0 is equal to zero, A_- equals zero at $\pi(\vec{q}) > 0$ and A_+ is equal to zero at $\pi(\vec{q}) < 0$; $\sigma_1^{(1)}$ and pair CF are given by the formulae (5.3), (5.5).

Figure 2 exhibits the pair CF (5.9) of the diluted system on the plane square lattice ($z = 4$). In the figure 3 one can observe that presence of many relaxation times in dynamic pair CF of the diluted system can obviously be seen at small temperatures and frequencies. Abscissas of maxima of $-\text{Im } m_{11}^{(2)}(\vec{0}, \omega)$ approximately equal inverse relaxation times: $\omega_0 \approx 1/\tau_1^0$, $\omega_+ \approx 1/\tau_+(\vec{0})$. Specific temperature dependence of $m_{11}^{(2)}(\vec{0}, \omega)$ is connected with nonmonotonous behaviour of $\tau_+(\vec{0})$.

6. Comparison to experimental data

One can see that TCA reflects essential properties of the diluted system. It seems to us that quality of the approximation is even higher for the system without noninteracting impurities ($K_{\alpha\beta} \neq 0$) because such "effective field" theories are more appropriate in the case of small fluctuations in the system considered. Obtained within TCA characteristics may be used in order to describe a variety of experimental data on partially deuterated ferroelectrics with hydrogen bonds and alloys of magnets. For example, good fit of TCA results for the ideal (one-sort) system to experimental data on the ferroelectric dispersion in pseudo-one-dimensional ($z = 2$) ferroelectrics CsH_2PO_4 and its deuterated analog CsD_2PO_4 has been reached in the ref. [10,11]. The many-sort theory developed here is applicable to partially deuterated compound $-Cs(H_{1-x}D_x)_2PO_4$. We obtain a good agreement of TCA results for annealed model with those of dielectric measurements [12,13] (figure 4). Fit of the quenched model (figure 5) to experimental data is not so obvious. The model parameters are shown in the table 1. More information about an actual type of the system can be delivered by measurement in the low-temperature region, where significant differences between behaviours of the annealed and quenched models exist, and by the wavevector-dependent experiments (e.g., neutron scattering).

Acknowledgements

The research described in publication was made possible in part by Grant No K6A100 from the Joint Fund of the Government of Ukraine and International Science Foundation.

References

- [1] Glauber R.J. Time-dependent statistics of the Ising model. // J. Math. Phys., 1963, vol. 4, No 2, p. 294-307.
- [2] Levitskii R.R., Sorokov S.I., Sokolovskii R.O. Correlation functions of quenched and annealed Ising systems. // Cond. Matt. Phys., 1994, No 5, p. 81-104 (in Ukrainian).
- [3] Suzuki M., Kubo R. Dynamics of the Ising model near the critical point. I. // J. Phys. Soc. Japan, 1968, vol. 24, No 1, p. 51-60.

- [4] Hilhorst H.J., Suzuki M., Felderhof B.U. Kinetics of the stochastic Ising chain in a two-flip model. // *Physica*, 1972, vol. 60, p. 199-222.
- [5] Calef D.F. Impurity dynamics in a one-dimensional chain. // *J. Stat. Phys.*, 1983, vol. 32, No 1, p. 81-94.
- [6] Yukhnovskii I.R., Levitskii R.R., Sorokov S.I. Thermodynamics and distribution functions of Ising model. Two-site cluster approximation. // Kiev, 1986, 33p. (Preprint / Acad. Sci. Ukr.SSR. Inst.Theor.Phys.; ITP-86-142R). (In Russian).
- [7] Sato H., Arrott A. Remarks on magnetically dilute systems. // *J. Phys. Chem. Solids*, 1959, vol. 10, p. 19-34.
- [8] Kaneyoshi T., Tamura I., Honmura R. Diluted Ising ferromagnet: Its physical properties. // *Phys. Rev. B*, 1984, vol. 29, p. 2769-2776.
- [9] Dattagupta S. Kinetics of ordering. // *Physica A*, 1993, vol. 194, p. 137-153.
- [10] Levitskii R.R., Grigas J., Zachek I.R., Mits Ye.V. Relaxational dynamics of quasi-one-dimensional CsD_2PO_4 -type ferroelectrics. // *Ferroelectrics*, 1986, vol. 67, p. 109-124.
- [11] Grigas J., Zachek I.R., Zaitseva P., Levitskii R.R. Ferroelectric dispersion in CsD_2PO_4 . // *Litovski fizicheski sbornik*, 1988, vol. 28, p. 486-498.
- [12] Deguchi K., Okaue E., Nakamura E. Effects of deuteration on the dielectric properties of ferroelectric CsH_2PO_4 . i. Static dielectric properties. // *J. Phys. Soc. Japan*, 1982, vol. 51, No 11, p. 3569-3574.
- [13] Deguchi K., Okaue E., Nakamura E. Effects of deuteration on the dielectric properties of ferroelectric CsH_2PO_4 . ii. Dynamic dielectric properties. // *J. Phys. Soc. Japan*, 1982, vol. 51, No 11, p. 3575-3582.

Appendix

Here we consider a system with the Hamiltonian

$$-\mathcal{H}/k_B T = \frac{1}{2} \sum_{i\alpha, j\beta} K_{i\alpha, j\beta} S_{i\alpha} S_{j\beta} + \frac{1}{2} \sum_{i\alpha, j\beta} J_{i\alpha, j\beta} S_{i\alpha} S_{j\beta} + \sum_{i\alpha} \Gamma_{i\alpha} S_{i\alpha}, \quad (\text{A1})$$

where $K_{i\alpha, j\beta}$, $J_{i\alpha, j\beta}$ are pair interaction parameters, $\Gamma_{i\alpha}$ external field acting on the spin of sort α on the site i . The interaction $K_{i\alpha, j\beta}$ is the same as in (2.1)-(2.3), but $J_{i\alpha, j\beta}$ is assumed to be of long-range type, i.e. not restricted to the nearest neighbour sites. The Hamiltonian (A1) can be written as

$$\mathcal{H} = {}^{MFA}H - \frac{k_B T}{2} \sum_{i\alpha, j\beta} J_{i\alpha, j\beta} \Delta^L m_{i\alpha}^{(1)} \Delta^L m_{j\beta}^{(1)}, \quad (\text{A2})$$

$${}^{MFA}H = H + \frac{k_B T}{2} \sum_{i\alpha, j\beta} J_{i\alpha, j\beta} {}^L m_{i\alpha}^{(1)} {}^L m_{j\beta}^{(1)},$$

$$\Delta^L m_{i\alpha}^{(1)} = S_{i\alpha} - {}^L m_{i\alpha}^{(1)},$$

where H is the Hamiltonian (2.1) in which the field κ is taken in the form

$$\kappa_{i\alpha} = \Gamma_{i\alpha} + \sum_{j\beta} J_{i\alpha, j\beta} {}^L m_{j\beta}^{(1)}. \quad (\text{A3})$$

MFA assumes

$${}^L m_{i\alpha}^{(1)} = \langle \langle S_{i\alpha} \rangle_{\mathcal{H}, t} \rangle_x \quad (\text{A4})$$

and ignores last term of (A2):

$$\mathcal{H} \longrightarrow^{MFA} H = H + \frac{k_B T}{2} \sum_{i\alpha, j\beta} J_{i\alpha, j\beta} {}^L m_{i\alpha}^{(1)} {}^L m_{j\beta}^{(1)}. \quad (\text{A5})$$

One can see that formalism of the section 3 is applicable to the system with the Hamiltonian (A5). The equation of motion has the same form

$$D_i \langle S_i \rangle_{\mathcal{H}, t} = \langle \tanh \beta \varepsilon_i \rangle_{\mathcal{H}, t}; \quad \varepsilon_i = \sum_j K_{ij} S_j + \kappa_i, \quad (\text{A6})$$

but now the field κ_i includes a molecular field (see (A3)) and thus couples the selfconsistency parameter ${}^L m_{i\alpha}^{(1)}$. Then one can find that

$${}^L m_{i\alpha}^{(1)} = m_{i\alpha}^{(1)}, \quad (\text{A7})$$

where $m_{i\alpha}^{(1)}$ is defined by the equation (3.13) and

$$\begin{aligned} {}^L m_{i\alpha, j\beta}^{(2)}(\omega) &\equiv \frac{\delta {}^L m_{i\alpha}^{(1)}(\omega)}{\delta \Gamma_{j\beta}(\omega)} = \sum_{l\gamma} \frac{\delta m_{i\alpha}^{(1)}(\omega)}{\delta \kappa_{l\gamma}(\omega)} \frac{\delta \kappa_{l\gamma}(\omega)}{\delta \Gamma_{j\beta}(\omega)} \\ &= \sum_{l\gamma} m_{i\alpha, l\gamma}^{(2)}(\omega) \left(\delta_{lj} \delta_{\gamma\beta} + \sum_{k\delta} J_{l\gamma, k\delta} {}^L m_{k\delta, j\beta}^{(2)}(\omega) \right). \end{aligned} \quad (\text{A8})$$

(A8) leads to the following expression for the structure factor

$$\left({}^L \hat{m}^{(2)}(\vec{q}, \omega) \right)^{-1} = \left(\hat{m}^{(2)}(\vec{q}, \omega) \right)^{-1} - \hat{J}(\vec{q}), \quad (\text{A9})$$

where $\hat{m}^{(2)}(\vec{q}, \omega)$ is defined by (3.14) and $\left(\hat{J}(\vec{q}) \right)_{\alpha\beta}$ is the Fourier-transform of $J_{i\alpha, j\beta}$.

РЕЛАКСАЦІЙНА ДИНАМІКА НЕВПОРЯДКОВАНОЇ МОДЕЛІ ІЗІНГА. НАБЛИЖЕННЯ ДВОЧАСТИНКОВОГО КЛАСТЕРА

Р.Р.Левицький, С.І.Сороков, Р.О.Соколовський

На основі кінетичного рівняння Глаубера досліджується спінова релаксація в моделі Ізінга з вузловим безладом. Обчислено динамічний структурний фактор моделі. Обговорюються ефекти, наявні в наближенні двочастинкового кластера і не присутні в наближенні молекулярного поля.

Отримані результати порівняні з діелектричними властивостями $Cs(H_{1-x}D_x)_2PO_4$.