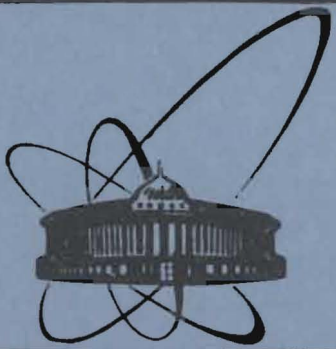

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STRUCTURAL GLASS IN THE TRANSVERSAL ISING MODEL WITH RANDOM COMPETING INTERACTIONS

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1. The solid solution $Rb_{1-x}(NH_4)_xH_2PO_4$ (RADP) of the order-disorder type ferroelectric (FE) RbH_2PO_4 (RDP) and antiferroelectric (AFE) $NH_4H_2PO_4$ (ADP) seems to be a structurally frustrated system like the conventional magnetic spin glass which is especially suitable for the experimental study of peculiarities of the glassy phase. Recently some experimental data on the phase diagram, susceptibility [1-8] and the dynamical behaviour [4,5] of this mixed crystal have been reported.

A theoretical estimation of the phase diagram based on the Ising pseudospin model for hydrogen bond systems within the cluster approximation has been given by Prelovsek and Blinc [6] and by Matsushita and Matsubara [7]. Unlike magnetic systems with Ising type coupling this approach takes into account the Slater rules for the protons and becomes, therefore, more complicated. In the work of Matsushita and Matsubara [7] the question of the appearance of a glassy phase remains open and the authors have suggested to map the system with ice-rule constraints onto a disordered Ising spin model with special interactions. Such a mapping for a nonrandom system has been recently investigated by Levstik et al. [8], too.

In this letter we consider the phase transitions in the transversal Ising model with random competing interactions within the mean-field approximation (MFA) and the virtual-crystal approximation (VCA) and discuss its relation to the solid solution RADP, especially, the appearance of the so-called structural glass (SG) phase.

2. The Hamiltonian of the pseudo-spin Ising model in a transverse field reads

$$H = -\Omega \sum_i S_i^x - (1/2) \sum_{ij} J_{ij} S_i^z S_j^z - \sum_i E_i S_i^z, \qquad (1)$$

where the coupling constants J_{ij} are random quantities with the known distribution function $P(J_{ij})$, and E_i denotes an external field. The thermal average of S_i^z can be calculated within MFA ($E_i = 0$) as

$$\langle S_i^z \rangle = m_i = (H_i^z / 2H_i) \tanh (H_i / 2kT), \qquad (2)$$

$$H_i^2 = \Omega^2 + (H_i^z)^2, \quad H_i^z = \sum_j J_{ij} m_j. \qquad (3)$$

For a sufficiently low temperature and small tunnelling constant Ω eq.(2) has a solution $m_i \neq 0$ which is in general inhomogeneous owing to the randomness of the J_{ij}. The solution m_i depending on the couplings J_{ij} represents itself a random field which may be described by its moments $\overline{m_i}$, $\overline{m_i m_j}$ etc., where the bars denote averages with $P(J_{ij})$.

To estimate the phase-transition temperature, let us investigate the stability of the paraphase ($m_i = 0$) with respect to a spin freezing. At first we consider the possibility of a phase with $\overline{m_i} \neq 0$ to appear. Note that $\overline{m_i}$ is in general site-dependent, e.g., in an antiferroelectric phase. Linearizing eq.(2) in m_i

$$m_i = A \sum_j J_{ij} m_j, \quad A = (1/2\Omega) \tanh(\Omega/2kT) \tag{4}$$

and averaging over J_{ij} within VCA, i.e., $\overline{J_{ij} m_j} \approx \overline{J_{ij}}\ \overline{m_j}$, after the Fourier transformation (assuming as usual $\overline{J_{ij}}$ to be translational invariant), $(1 - A\overline{J_q})\overline{m_q} = 0$, which yields the transition temperature T_c to a phase modulated with the wave vector q_0, $\overline{m_i} \sim \cos q_0 R_i$, we get:

$$kT_c = (\Omega/2) / \operatorname{artanh}(2\Omega/\overline{J}_{q_0}), \quad \overline{J}_{q_0} = \max \overline{J}_q. \tag{5}$$

To estimate the transition temperature to the SG phase with $\overline{m_i} = 0$ but $\overline{m_i^2} \neq 0$, we use a method [9] similar to an approach of Sherrington [10] in the mean-field theory of spin glasses. Squaring eq.(4) and averaging in the VCA with

$$\overline{J_{ik} J_{i\ell} m_k m_\ell} \approx \overline{J_{ik} J_{i\ell}}\ \overline{m_k m_\ell} \approx \overline{J_{ik} J_{i\ell}}\ \overline{m_k^2}\,\delta_{k\ell}$$

we obtain

$$(1 - A^2 J_2^2)\overline{m_k^2} = 0, \quad J_2^2 = \sum_j \overline{J_{ij}^2}. \tag{6}$$

In eq.(6) we have further supposed that $\overline{m_k^2}$ does not depend on the site. According to eq.(6) the paraphase becomes unstable with respect to a freezing into the SG phase at the temperature

$$kT_g = (\Omega/2)/\operatorname{artanh}(2\Omega/J_2). \tag{7}$$

Now let us apply the derived estimations to a system, where the sites of the model (1) are randomly occupied by atoms of the kind A or B. Then we get $\overline{J_{ij}} = x_A^2 J_{ij}^{AA} + 2x_A x_B J_{ij}^{AB} + x_B^2 J_{ij}^{BB}$, where x_A, x_B are the concentrations of the components. For simplicity we choose $J_{ij}^{AA} = -J_{ij}^{BB} > 0$, $J_{ij}^{AB} = 0$ and suppose a simple cubic lattice with the nearest neighbour interactions. This yields $\overline{J}_{q0} = J_0^{AA} |x_A - x_B|$, $J_0^{AA} = \sum_i J_{ij}^{AA}$ and $J_2^2 = (J_0^{AA})^2 (x_A^2 + x_B^2)/6$. The temperatures of the transition to the FE and AFE phase, respectively, and to the SG phase according to eqs. (5) and (7) are plotted in Fig.1.

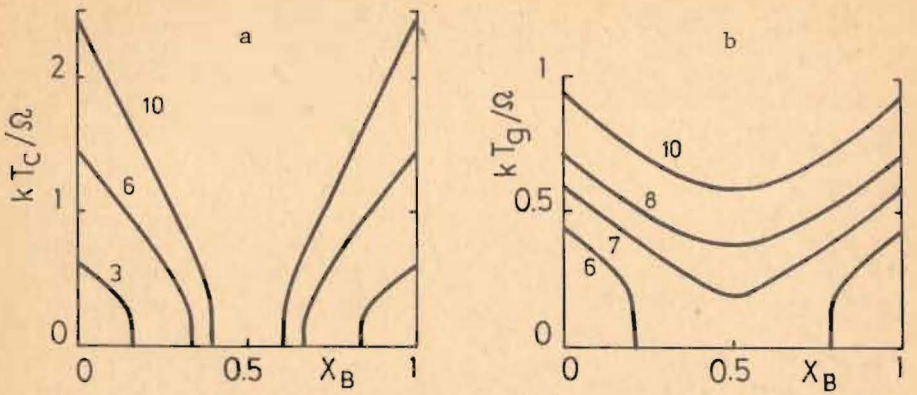


Fig.1. Transition temperatures T_c (a) and T_g (b), respectively, versus concentration x_B for different parameters J_0^{AA}/Ω.

The static susceptibility defined by $\chi_{ik} = (\partial m_i/\partial E_k)_{E_k=0}$ may be obtained from (2) if we substitute H_k^z by $H_k^z + E_k$:

$$\chi_{ik} = \chi^0(H_i^z)(\delta_{ik} + \sum_j J_{ij} \chi_{jk}), \tag{8}$$

$$\chi^0(H_i^z) = [(\Omega^2/2H_i) \tanh(H_i/2kT) + ((H_i^z)^2/4kT)(1 - \tanh^2(H_i/2kT))]/H_i^2. \tag{9}$$

An average of this equation within VCA yields after Fourier transformation

$$\overline{\chi}_q = \overline{\chi^0(H_i^z)}\,(1 - \overline{J}_q\,\overline{\chi^0(H_i^z)})^{-1}. \tag{10}$$

To discuss the susceptibility, we restrict ourselves to the case of a glass phase: $\overline{m_j} = 0$, $\overline{m_i^2} = q \neq 0$ and $\overline{J_q} = 0$. With the approximations $\overline{f((\sum_j J_{ij} m_j)^2)} \approx f(\overline{(\sum_j J_{ij} m_j)^2}) \approx f(J_2^2 q)$ where f is an arbitrary function, after averaging the squared eq.(2) we get

$$q = (1/4)\tanh^2((\Omega^2 + J_2^2 q)^{1/2}/2kT) - \Omega^2/J_2^2, \tag{11}$$

governing the temperature dependence $q(T)$. The average of eq.(9) yields

$$\overline{\chi^0(H_i^z)} = \chi^0(H_z), \quad H_z = (J_2^2 q)^{1/2}. \tag{12}$$

The solution $\bar{\chi}$ of the system of equations (10)-(12) is plotted in Fig.2.

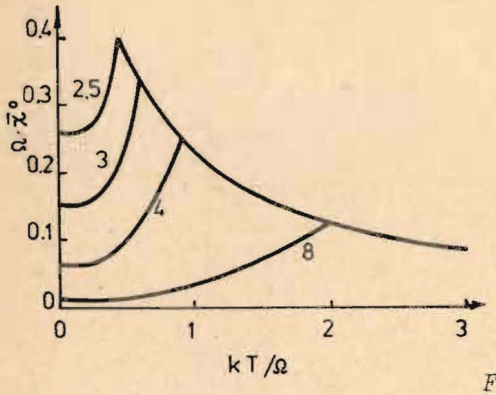


Fig.2. Susceptibility versus temperature at the concentration $x_B = 0.5$ for different parameters J_2/Ω.

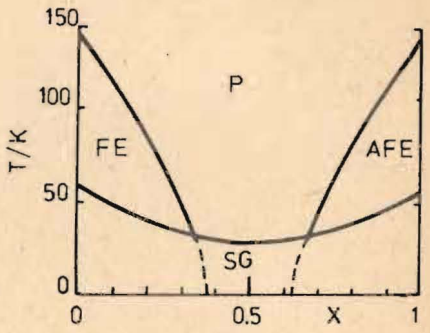
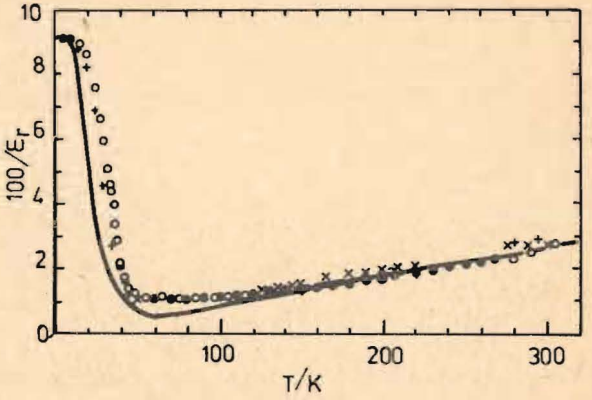


Fig.3. Calculated phase diagram for RADP showing the stability limits of the paraphase (P) with respect to a freezing into the FE, AFE and SG phases.

3. Let us now apply the presented model to RADP. To obtain a phase diagram like the experimental one the nonrandom tunneling constant has been chosen as $\Omega/k = 73$ K. Then $T_c = 146$ K for RDP yields $J_0^{RDP}/k = 596$ K and for ADP we assume $J_0^{ADP} = -J_0^{RDP}$. The couplings between ADP and RDP sites are supposed to be zero. The transition temperatures for this example following from (5) and (7) are given in Fig.3. For the concentration $x = 0.35$ a glass transition temperature T_g about 30 K is obtained in agreement with experiment, but T_c is only a little bit lower. The calculated phase diagram predicts a concentration range $x = 0.35$-0.65 where a glass transition occurs in comparison to experimental results with $x \approx 0.2$-0.8.

Recently, the experiments on deuterated RADP have been published by Schmidt et al.[3]. The parameters of our model depend in general on the deuteration fraction D. In case of linear interpolation we get $\Omega(D) = (1-D)\Omega(0) + D\Omega(1)$ and an analogous formula for $J_0(D)$. The coupling constants for $D = 1$ may be obtained from the transition temperatures $T_c = 218$ K for DRDP and $T_c = 242$ K for DADP[11]. Assuming further $\Omega(D=1) = 0$ and the coupling between sites of different kind $J_0^{AB}(D) = (J_0^{AA}(D) + J_0^{BB}(D))/2$, we finally find for the reported concentrations $D = 0.71$ and $x = 0.48$: $|\bar{J}_0|/k \ll \Omega/k = 21$ K and $J_2/k \approx 240$ K. These parameters yield the glass transition temperature $T_g \approx 60$ K in good agreement with experiment, although the peak of the measured

Fig.4. Inverse dielectric permittivity (full line) versus temperature for deuterated RADP in comparison with the experimental results obtained by Schmidt et al. (1984).



susceptibility is rather broad and, therefore, the transition temperature is not well defined experimentally. Since for our example $|\bar{J}_0| \ll J_2$, we expect the appearance of a pure glassy state ($\overline{m_i} = 0$) and the susceptibility can be calculated from (10)-(12).

To compare our results with experiment, we use the formula $\epsilon_r = \epsilon_0 + C\Omega\bar{\chi}^0$ for the relative permittivity, where ϵ_0 denotes a background susceptibility due to further ionic and electronic polarizability and C has the meaning of a Curie-Weiss constant. With the fit parameters $\epsilon_0 = 5.5$ and $C = 1900$ the function ϵ_r^{-1} is presented in Fig.4 in comparison with the experimental curve of Schmidt et al.[3]. The largest deviations of both curves at the transition temperature are caused by the sharp cusp behaviour of our model as compared to the observed broad susceptibility peak.

4. In conclusion, the random transversal Ising model treated within the MFA and VCA describes some features of the experiments on the solid solution RADP at least qualitatively. The consideration of the tunnelling motion of the protons seems to us to be important especially for mean concentrations ($x \approx 0.5$) since in this case the effective interaction strength J_2 and correspondingly the glass transition temperature T_g become comparable with the tunnelling constant.

A more realistic description of RADP requires, presumably, a cluster approximation considering the Slater rules as well as the tunnelling. One also should take into account the hydrogen bonds due to the NH_4 groups since the freezing of the hydrogen motion in these groups is important for the transition to the glassy state as has been reported by Slak et al.[5]. The latter experiments also suggest that there is no true equilibrium phase transition to the glass phase, and therefore, our calculation of the transition temperature T_g should be regarded as some hints where dynamical peculiarities may be expected.

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СООБЩЕНИЯ, КРАТКИЕ СООБЩЕНИЯ, ПРЕПРИНТЫ И СБОРНИКИ ТРУДОВ КОНФЕРЕНЦИЙ, ИЗДАВАЕМЫЕ ОБЪЕДИНЕННЫМ ИНСТИТУТОМ ЯДЕРНЫХ ИССЛЕДОВАНИЙ, ЯВЛЯЮТСЯ ОФИЦИАЛЬНЫМИ ПУБЛИКАЦИЯМИ.

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Савин И.А., Смирнов Г.И. В сб."Краткие сообщения ОИЯИ", № 2-84, Дубна, 1984, с.3.

Аксенов В.Л., Бобет М., Плакида Н.М. E17-85-163

Структурное стекло в модели Изинга в поперечном поле со случайными конкурирующими взаимодействиями

В приближении среднего поля и в приближении виртуального кристалла вычислены фазовая диаграмма и статическая восприимчивость модели Изинга в поперечном поле со случайными конкурирующими взаимодействиями. Обсуждается связь модели с твердым раствором $Rb_{1-x}(NH_4)_xH_2PO_4$, в котором предполагается случайность по узлам решетки.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1985

Aksenov V.L., Bobeth M., Plakida N.M. E17-85-163

Structural Glass in the Transversal Ising Model with Random Competing Interactions

Phase diagram and static susceptibility of the transversal Ising model with random competing interactions are calculated within the mean-field and virtual-crystal approximations. The relation of this model to the solid solution $Rb_{1-x}(NH_4)_xH_2PO_4$ is discussed assuming this system to be site-random.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1985