ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА

E17 - 10286

J.Schreiber

PSEUDO-SPIN MOTION IN SOLID SOLUTIONS OF THE TYPE $K(H_{1-x}D_x)_2PO_4$

1976

E17 - 10286

J.Schreiber

PSEUDO-SPIN MOTION IN SOLID SOLUTIONS OF THE TYPE $K(H_{1-x}D_x)_2PO_4$

Submitted to "physica status solidi"

Шрейбер Ю.

E17 - 10286

Псевдоспиновое движение в твердых растворах типа $K(H_{1-x}D_x)_2PO_4$

Исследуется эффект дейтеризации в рамках псевдоспиновой модели сегнетоэлектриков типа KDP с учётом изменения константы туннелирования Γ_i, дипольного момента μ_i и констант взаимодействия $I_{ij} \sim \mu_i t_{ij} \mu_j$. Вычислена диэлектрическая восприимчивость с помощью линеаризованного уравнения Блоха и метода когерентного потенциала. Обсуждена концентрационная зависимость точки Кюри, поляризации, интенсивности рамановского рассеяния и статистической восприимчивости и их связь с экспериментальными данными.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Препринт Объединенного института ядерных исследований
Дубна 1976

Schreiber J.

E17 - 10286

Pseudo-Spin Motion in Solid Solutions of the Type $K(H_{1-x}D_x)_2PO_4$

Within the pseudo-spin model for ferroelectric solid solutions of the type $K(H_{1-x}D_x)_2PO_4$ which includes occupation dependent tunneling frequencies Γ_i, dipole moments μ_i, and interactions $I_{ij} \sim \mu_i t_{ij} \mu_j$, the dielectric susceptibility is obtained by a linearized Bloch equation.

The single site and the Bethe-Peierls version of CPA was used to solve the disorder problem. Using a suitable set of ingoing parameters the Curie temperature, the spontaneous polarization, the intensity of Raman scattering, and the static susceptibility are calculated for various deuterium concentrations x and discussed in relation to experiments.

Preprint of the Joint Institute for Nuclear Research
Dubna 1976

1. *Introduction*

There are mainly two reasons for the investigation of solid solution of the KDP type ferroelectrics. Since the characteristical quantities of these systems are strongly concentration dependent, a quite good test for several theoretical models and applied approximations can be made [1]. On the other hand, it is of fundamental interest to understand the influence of disorder on the ferroelectric properties analogously to disordered magnetic systems [2-4]. For instance, it is known that the Raman spectra for undeuterated KDP have a characteristic peak which, however, does not appear in the complete deuterated case (see ref. [5]). In this connection several interesting questions arise concerning the mixed system, e.g., how do the intensity and the frequency of the mentioned peak change with increasing concentration of deuterium. So far only preliminary experimental results are available [6] which indicate that the frequency of the underdamped soft mode below the Curie temperature T_c increasing deuteration, although its intensity is found decreasing. Within the pseudo-spin model (ref. [5]) the measured peak in the KDP system can be attributed to collective pseudo-spin excitations. The resonant character of these modes disappears on deuteration, i.e., only local excitations exist which do not contribute to the Raman scattering. Therefore, the question arises whether or not the transfer from collective to nonresonant local modes shows a drastic change in the peak intensity of mixed systems at a critical concentration [7] similar to the percolation concentration [8] or to that in the case of dilute ferromagnets [9].

In this paper we give a theoretical explanation, first of all, for dynamical properties of deuterated KDP in the framework of the pseudo-spin model. The Hamiltonian has the form

$$H = -\sum_i \Gamma_i S_i^x - \frac{1}{2}\sum_{ij} I_{ij} S_i^z S_j^z - 2\sum_i \mu_i E_i^z(t) S_i^z, \qquad (1)$$

where phonons and phonon-pseudo-spin coupling are explicitly neglected while we use renormalized tunneling frequency and pseudo-spin interactions (see refs.[4,10]). In mixed systems $K(H_{1-x}D_x)_2PO_4$ the parameters in (1) depend on the occupation of the bonds i by protons H or deuterons D which have been assumed to be homogeneously and randomly distributed. The increase of the mass by the replacement $H \to D$ causes a rapid decrease of the tunneling frequency, i.e., $\Gamma_i^H \gg \Gamma_i^D \approx 0$ (cf.[5,10]). Using the molecular field approximation (MF)[11] the change of Γ_i alone explains the isotopical effect on the Curie temperature in deuterated KDP qualitatively. On the other hand, also the inetraction between the bonds will depend on the type of bonds. Following Holakovsky[12], Kopsky[13], and Vaks et al.[1] we adopt only such interactions which are proportional to the dipole moments, i.e., $I_{ij} \sim \mu_i t_{ij} \mu_j$ (t_{ij} is a geometrical term).

The static properties of $K(H_{1-x}D_x)_2PO_4$ were investigated by MF and cluster approximations[1,11-13]. Using the virtual crystal approach, Blinc et al.[14] have discussed the pseudo-spin motion in mixed proton-deuteron systems including coupled phonon mode.

Here we describe the pseudo-spin motion with the help of a Bloch equation[5], , i.e., the equation of motion is derived within the random phase approximation (RPA), where the static properties have been calculated by the MF, and have added phenomenological local relaxation terms (Sect. 2). We have treated the disorder problem on the basis of the coherent potential approximation (CPA) in the single site (cf.[15]) and the Bethe-Peierls (cf.[16]) version (Sect 3). In agreement with experimental hints[6] it was found (Sect. 3.2) that the pseudo-spin wave peak in the Raman spectrum at $T < T_c$ shifts to higher energies with increasing the concentration of deuterium. The intensity of the peak rapidly decreases for small deuterium concentrations, however, no critical concentration is found, i.e., the peak only asymptotically vanishes as $x \to 1$ For the explanation of the rapid decrease of the intensity we have to take into account not only the elastic disorder scattering. The essential effect is due to the inelastic scattering of pseudo-spin waves at deuteron bonds.

We have also calculated the static susceptibility (Sect. 3.3). Within our approximation we could not find stable solution in the neighbourhood of T_c. To overcome this difficulty one has to include thermal and structural fluctuations in a better way (cf.[17]) or to perform a self-consistent RPA calculation (cf.[18]).

2. CPA for Pseudo-Spin Motion

The pseudo-spin motion in the system (1) may be described by the Bloch equation (cf[5]), where one considers the procession of pseudo-spins around the molecular field $\vec{H}_i(t)$ in RPA. Damping and pseudo-spin motion along the molecular field are included by adding phenomenological relaxation terms to the RPA equations of motion. The relaxation times T_1 and T_2 characterize the relaxation of spin components parallel and perpendicular to the instantaneous molecular field direction. In the sense of linear response to an external field $E_i^z(t) = E_i^z e^{i\omega t}$ we suppose

$$\vec{S}_i(t) = \vec{\bar{S}}_i + \delta\vec{S}_i e^{i\omega t}, \quad \vec{H}_i(t) = \vec{H}_i + \Delta\vec{H}_i e^{i\omega t}, \qquad (2)$$

where the expectation value of the pseudo-spin $\vec{S}_i$ is obtained as

$$\vec{\bar{S}}_i = \frac{1}{2}\frac{\vec{H}_i}{H_i}\tanh\left(\frac{H_i}{2kT}\right), \quad \vec{H}_i = \left(\Gamma_i, 0, \sum_j I_{ij}\overline{S_j^z}\right). \qquad (3)$$

The linearization of the Bloch equation yields the time dependent deviation $\delta P_i(t)$ from the spontaneous polarization P_i

$$\delta P_i(t) = 2N\mu_i \delta \bar{S}_i^z(t) = \sum_j \chi_{ij}^{zz}(\omega) E_j^z(t), \qquad (4)$$

where the dielectric susceptibility $\vec{\chi}_{ij}(\omega)$ is determined by the equation

$$\vec{\chi}_{ij} = (\chi_{ij}^{xz}, \chi_{ij}^{yz}, \chi_{ij}^{zz}) = \vec{g}_i(\omega)\delta_{ij} + \overleftrightarrow{g}_i \sum_k t_{ik}\vec{\chi}_{kj} . \qquad (5)$$

The single particle like suscebtibility $\vec{g}_i(\omega)$ (locator) was found to be a function of Γ_i, μ_i, $\vec{S}_i$, T_{1i}, and T_{2i} (see the Appendix). The 3 x 3 matrix $\overleftrightarrow{g}_i$ has only a non-zero third column which is equal to the vector $\vec{g}_i$. For the random system we have to average $\delta P_i(t)$ over all configurations for a given concentration of deuterium. Here we describe three approximations.

2.1. Averaged Locator Approach (ALA)

Using the zero order approximation in a perturbation sense, i.e., $\langle \overleftrightarrow{g}_i \vec{\chi}_{kj} \rangle \approx \langle \overleftrightarrow{g}_i \rangle \langle \vec{\chi}_{kj} \rangle$, we obtain for the Fourier transform of $\langle \vec{\chi}_{ij} \rangle$

$$\vec{\chi}(\vec{k},\omega) = (1 - \langle \overleftrightarrow{g}_i \rangle t_{\vec{k}})^{-1} \langle \overleftrightarrow{g}_i(\omega) \rangle . \qquad (6)$$

The quantity $\langle \vec{g}_i(\omega) \rangle$ may be calculated within an extended molecular field approximation (cf. /3/), i.e, we neglect the structural fluctuations in the neighbourhood of bond i when calculating the molecular field $\vec{H}_i$ (for $j \neq i$ we replace $\mu_j \vec{S}_j$ by $\langle \mu_j \vec{S}_j \rangle$). Then we have easily $\langle \vec{g}_i \rangle = (1-x)\vec{g}_H + x\vec{g}_D$, where $\vec{g}_H$ is the corresponding locator when at the bond i a proton appears.

2.2. Single Site CPA (SCPA)

In the usual CPA sense we try to find a solution of Eq. (5) in the form

$$\langle \vec{\chi}_{ij} \rangle = \vec{\sigma}_{ij} + \sum_{k,\ell} \sigma_{ik} t_{k\ell} \langle \vec{\chi}_{\ell j} \rangle = \vec{\chi}^c_{ij} , \qquad (7)$$

where $\vec{\sigma}_{ij}$ describes an effective medium and will be determined self-concistently. In the single site approximation it is assumed that $\vec{\sigma}_{ij}$ is a local quantity, i.e., $\vec{\sigma}_{ij} = \vec{\sigma}\delta_{ij}$. The self-consistent equation may then be obtained by the more phenomenological requirement

$$\langle \vec{\chi}_{ii} \rangle_i = (1-x)\vec{\chi}_H + x\vec{\chi}_D = \vec{\chi}^c_{ii} , \qquad (8)$$

where the site i is embedded in the effective medium, i.e., for the determination of $\vec{\chi}_{ii}$ according to Eq. (5) $\vec{g}_j(\omega)$ is replaced by $\vec{\sigma}$ for all $j \neq i$. The structural average $\langle \ldots \rangle_i$ is taken over the two occupational states of the bond i. Carrying out detailed calculations according to Shiba's approach /15/ and performing the matrix operations, we have found the self consistent equation for the z-component of $\vec{\sigma}$ in the form

$$\sigma^z(\omega) = \langle g_i^z(\omega) \rangle - (g_H^z(\omega) - \sigma^z(\omega)) F(\omega) (g_D^z(\omega) - \sigma^z(\omega)) , \qquad (9)$$

where

$$F(\omega) = 1/N \sum_{\vec{k}} t_{\vec{k}} / (1 - \sigma^z(\omega) t_{\vec{k}}) = \int d\omega' \rho_0(\omega') \omega' / (1 - \sigma^z(\omega)\omega'), \qquad (10)$$

and $\rho_0(\omega) = \sum_{\vec{k}} \delta(\omega - t_{\vec{k}})$. $\sigma^x(\omega)$ and $\sigma^y(\omega)$ are simple functions of $\sigma^z(\omega)$. We note that the same equation for determination of $\vec{\sigma}(\omega)$ follows if one uses the multiple scattering approach (cf. /19/) where the used approximation can be better estimated.

Since the real structure of KDP systems is quite complicated and also a long range interaction exists, it is too difficult to take the sum over $\vec{k}$ in an exact way. For principle investigations one either considers one dimensional systems /20/ or

uses simple approximations for $\rho_0(\omega)$. That means, the disorder effect is studied in a fictive system which has a simple topological structure and an approximated interaction, however, the ferroelectric model is conserved mainly. On the other hand, it can be supposed that long wave lenght properties like Raman scattering intensity are not very sensitive to structural details. In this sense here we use a model density of pseudo-spin states of the form

$$\rho_0(\omega) = 2/\pi t_0 (t_0^2 - \omega^2)^{1/2} \quad . \tag{11}$$

When calculating the quantities $\langle g_i^z \rangle$, g_H^z and g_D^z, we also use the extended molecular field approximation (see Sect. 2.1) in agreement with the single site approximation of the CPA. Numerical results for $\vec{\chi}(\vec{k}, \omega)$ are discussed in Sect. 3.

2.3. Bethe-Peierls Approach (BPA)

To include structural fluctuations in the environment of the bond i, we apply the improved CPA version proposed by Brouers et al. /16/. The effective locator $\vec{\sigma}$ is here also supposed to be site diagonal and the CPA requirement is similar to Eq. (8). In contrast to the simple single site CPA $\vec{\chi}_{ii}$ is calculated for a nearest-neighbour cluster surrounded by the effective medium. Thereby, in a first approximation all kinds of correlations between the nearest-neighbours (NN) are neglected, e.g., in the expansion of the diagonal self-energy only the first renormalized term is considered according to the Bethe lattice like approximation /16/. Therefore $\vec{\chi}_{ii}$ depends on the occupation of the bond i and only on the number of protons or deuterons in NN bonds. Keeping the assumption of a noncorrelation distribution of deuterons, the configuration average of $\vec{\chi}_{ii}$ can be written as

$$\langle \vec{\chi}_{ii} \rangle = \sum_{n=0}^{Z} \frac{Z!(1-x)^n}{(Z-n)!\,n!} x^{Z-n} \{(1-x)\vec{\chi}_H^n + x\vec{\chi}_D^n\} \, , \tag{12}$$

where n denotes the number of protons in the NN sphere. The self-consistent equation for the z-component of $\vec{\sigma}$ reads now

$$\sigma^z = \sum_{n=0}^{N} \frac{Z!(1-x)^n}{(Z-n)!\,n!} x^{Z-n} \{(1-x)\sigma_H(n) + x\sigma_D(n)\},$$

$$\sigma_H(n) = g_H^z(n) / \{1 + F(\omega)[\sigma^z - g_H^z(n)(\frac{n}{Z} e_{HH}^n + (1 - \frac{n}{Z}) e_{DH}^n)]\},$$

$$e_{HH}^n = [1 + (\sigma^z - g_{HH}^z(n)) F_1(\omega)]^{-1} \, ,$$

$$F_1(\omega) = F(\omega)[Z\,(1 + \sigma^z F(\omega))]^{-1} \quad . \tag{13}$$

For the evaluation of g_H^z, $g_{HH}^z(n)$, and similar quantities (locators for the central and for an NN bond in the case of a certain occupation) an approximation beyond the homogeneous MF (see Sect. 2.1) is used. According to the BPA, the static expectation values of the pseudo-spins are self-consistently calculated for a special configuration of the considered NN cluster, where the effective field of the environment is structurally averaged and is homogeneous. Correlation between NN bonds are neglected, i.e., for an NN of bond i all other bonds, excluding the central bond, belong to the effective or structure averaged medium. Thus, e.g., $g_{HD}^z(n)$ denotes g_j^z for an NN bond occupied by a proton while the central bond is occupied by a deuteron, furthermore in this case the proton number at the NN bonds is n. Numerical calculations are here more extensive, however, could be done without difficulties (see Sect. 3).

3. *Results and Discussion*

3.1. Polarization and Curie Temperature

At first we recall the MF results for the spontaneous polarization $P_{T=0}(x)$ and the Curie temperatire $T_c(x)$ for a special set of the following parameters: Γ^H/t_0, kT_c^H/t_0, kT_c^D/t_0, μ_D/μ_H, where the relation

$$2\Gamma^H/t_0 = \tanh\left(\frac{\Gamma^H}{2kT_c^H}\right) \quad \text{and} \quad kT_c^D/t_0 = 0.25(\mu_D/\mu_H)^2 \qquad (14)$$

holds. The direct measurable quantities T_c^H and T_c^D are so far well determined. According to several experimental data the relation T_c^D/T_c^H varies from 1.73 /5/ and 1.74 /21/ up to 1.85 /1/. The relation μ_D/μ_H cannot be found directly (cf. /12,13/) and there are different estimations - $\mu_D/\mu_H = 1.33$ /1/ and $\mu_D/\mu_H = 1.24$ /21/. Here we use the experimental results obtained by Frenzel et al., i.e., $kT_c^H/t_0 = 0.22$, $kT_c^D/t_0 = 0.38$, $\Gamma^H/t_0 = 0.29$, and $\mu_D/\mu_H = 1.24$ (case I). The $T_c(x)$ and $P_{T=0}(x)$ curve for these parameters (Fig. 1) are compared with the theoretical case, where $\mu_D/\mu_H = 1$, , $kT_c^H/t_0 = 0.15$, , $kT_c^D/t_0 = 0.25$, and $\Gamma^H/t_0 = 0.46$ (case II). It is seen that the MF, including the change of interaction due to deuteration, yields $T_c(x)$ in relatively good agreement with experimental data (see /1/). Also the relation $(P_{T=0}(1)-P_{T=0}(0))/P_{T=0}(0)=0.47$ is not far from the experimental value (about 0.20 - cf /13/) in contrast to case II (1.63).

3.2. Raman Scattering

The intensity $I(\omega)$ of the Raman scattering, reflecting the polarization fluctuations is due to the fluc-

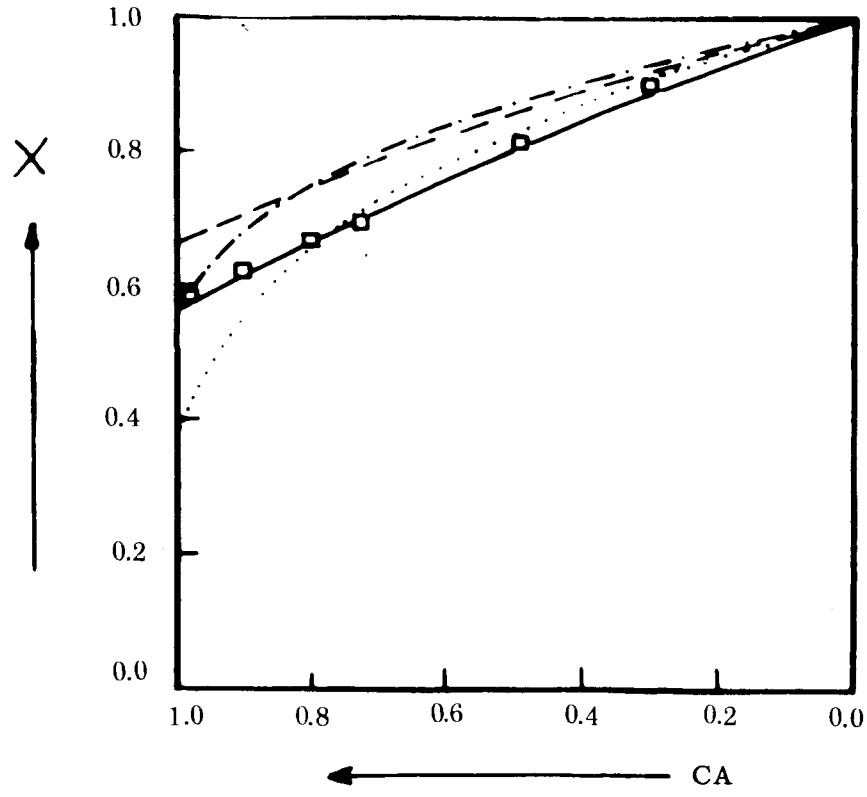

Fig. 1. Curie temperature $X = T_c(x)/T_c^D$ (—— $\mu_D/\mu_H=1.24$, $\Gamma^H/t_0=0.29$; -·-·- $\mu_D/\mu_H=1$, $\Gamma^H/t_0=0.46$) *and spontaneous polarization at* $T=0$ $X=P_{T=0}(x)/P_{T=0}^D$ (- - - $\mu_D/\mu_H=1.24$; ... $\mu_D/\mu_H=1$) *upon proton concentration* $CA=1-x$. *Experimental data* □ *are from Ref. 14 in* /1/.

tuation-dissipation theorem related to the imaginary part of the complex dynamic susceptibility (cf /5/). Concerning polarization fluctuation in the z-direction we have

$$I(\omega) \sim (e^{\omega/kT}/(e^{\omega/kT}-1))\mathrm{Im}\chi^{zz}(\vec{k}\to 0,\omega), \qquad (15)$$

where according to Eq. (7), (9), and (13) one may write

$$\chi^{zz}(\omega, \vec{k} \to 0) = \sigma^{z}(\omega)/(1 - \sigma^{z}(\omega)\, t_0). \qquad (16)$$

Numerical results are shown in Fig. 2 (case II) and Fig. 3 (case I) for $T<T_c$. The choice of the relaxation times - we have distinguished between proton (T_1^H, T_2^H) and deuteron relaxation (T_1^D) - is essentially arbitrary. T_1 influences particularly the width of the central peak around $\omega = 0$ and T_2 determines in general the width of the pseudo-spin wave peak (cf. Figs. 2 and 3).

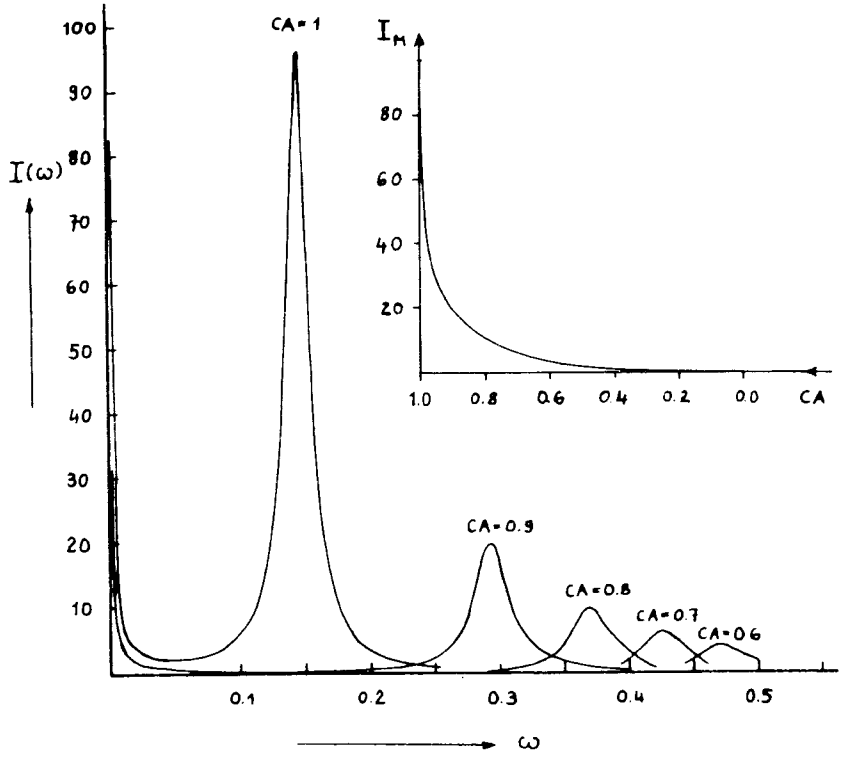

Fig. 2. Intensity of Raman scattering $I(\omega)$ *according to SCPA for various* $CA = 1-x$ *and* $kT/t_0 = 0.12$. I_M *is the amplitude of the pseudo-spin wave peak.* $\Gamma^H/t_0 = 0.46$, $\mu_D/\mu_H = 1$, $T_1^H = 0.002$, $T_2^H = 0.02$, *and* $T_1^D = 0.02$.

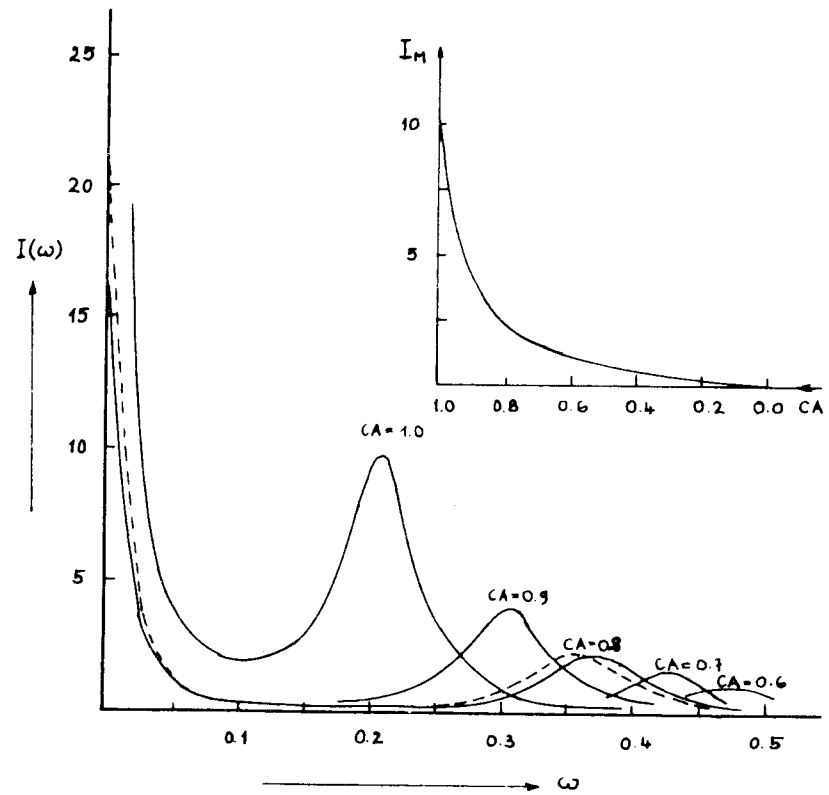

Fig. 3. $I(\omega)$ *according to SCPA* (——) *and BPA* (– – –) *for various* $CA = 1 - x$ *and* $kT/t_0 = 0.20$. I_M - *see Fig. 2.* $\Gamma^H/t_0 = 0.29$, $\mu_D/\mu_H = 1.24$, $T_1^H = 0.02$, $T_2^H = 0.04$ *and* $T_1^D = 0.04$.

In agreement with experimental hints /6/ the peak of the pseudo-spin wave mode is shifted to higher energies and the intensity rapidly decreases (Figs. 2 and 3) with increasing deuterium concentration. In the pure KDP case ($x=0$) the position of this peak is proportional to the polarization at $T < T_c$. The results show that also in the mixed system the frequency $\omega_s(x)$ of the pseudo-spin wave is related to the spontaneous polarization $P(x)$ because this quantity shows the same concentration dependence as $\omega_s(x)$ (see Figs. 1,2 and 3).

The falling off of the amplitude $I_M(x)$ (Figs. 2 and 3) is caused by the elastic and also inelastic scattering of pseudo-spin waves at randomly distributed deuteron scatterers. As spin excitations at energy $\omega \sim \omega_s$ are strongly absorbed at deuteron bonds the spin wave excitations are also absorbed due to their collective character. This is the reason why at small deuterium concentrations $I_M(x)$ decreases so rapidly, but the width of the peak does not increase adequately which would be expected from elastic disorder scattering only. In spite of the possibility that for concentrations $x > x_{cr}$ (about the percolation concentration) the excitations at proton bonds become localized, these excitations still contribute to the polarization fluctuations in the z-direction as the molecular field does not point along the z-direction.

The localization of these excitations may effect therefore only the damping, i.e., the width of the peak. However this effect is small since the width of the peak and the $I_M(x)$ curve give no indication of any critical concentrations. Perhaps, x_{cr}, if it exists may be detected by neutron scattering experiments. In Fig. 3 we have also plotted the results for the BPA. The qualitative behaviour of $I(\omega)$ is the same, even the quantitative corrections are small. The ALA yields also quantitative similar results, although quantitative deviations are larger, e.g., ALA does not show such a marked falling off at small x.

3.3 Static Susceptibility

In the critical region the used approximation becomes important. According to the ALA, $\chi^{-1}(T) = \chi^{zz}(\omega \to 0, \vec{k} \to 0)^{-1}$ is always real and positive and zero at $T = T_c$. However, the SCPA yields an instable region around T_c (see Fig. 4), i.e., $\chi(T \approx T_c)$ is complex and the real part becomes negative. That means, the considered disorder scattering leads to a tendency to destroy the long range order as early as predicted by the MF. Working therefore within a self-consistent theory also for the determination of $\vec{S}_i$ (see /18/) the instability in $\chi(T)$ could be cancell-

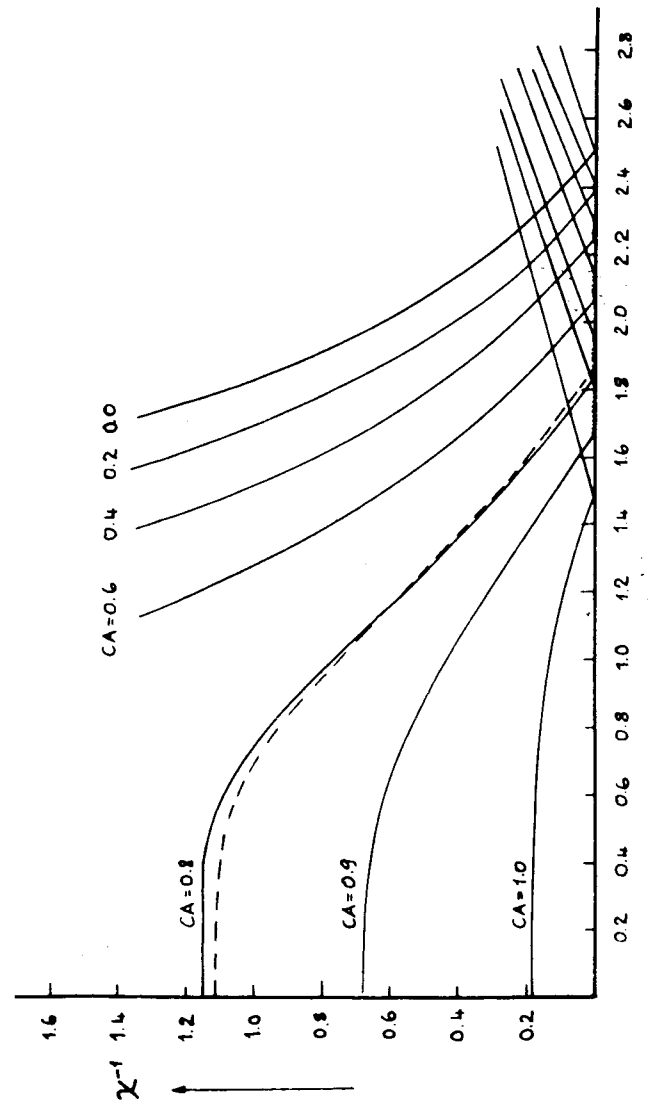

Fig. 4. Reciprocal susceptibility $\chi^{-1}(T)$ within SCPA (——) and BPA (– – –) upon temperature kT/t_0 for various CA = 1 – x. $\Gamma^H/t_0 = 0.46$, $\mu_D/\mu_H = 1$, and $t_0 = ZJ$.

ed. On the other hand, including environment effects with the BPA the MF-RPA solution becomes stabilized partially (see Fig. 4, curve x=0.2). Analogously to structural fluctuations it may be expected that also thermodynamical fluctuations stabilize the MF-RPA solution, This was confirmed by a simple approach, where a stochastic local field was introduced which fluctuates around the molecular field according to a model distribution function. This field and the structural disorder were commonly treated within the SCPA. A reduction of the unstable region was found. To improve this consideration we have to find a self-consistent condition for construction of the stochastic field distribution, possibly, in a way similar to the lines approach [17].

Acknowledgements

The author would like to express his thanks to Dr. R.Ferchmin for stimulation of this work, for many valuable discussions in connection with the used methods and approximation, and for his endeavour to connect this theoretical work with experiments. I am very grateful to Doc. B.Hilczer for valuable comments and stimulation of suitable experiments. The author is furthermore indebted to Dr. H.Brater, Dr. J.Kowalski, Dr. A.Popialewicz, Prof. K.Elk, and Dr. N.M.Plakida for interesting discussions.

Appendix

It is seen from Eqs. (9), (13), and (16) that only the z-component of $\vec{g}_i$ is necessary for detailed calculations. According to [5] we find the following expression for g_i^z

$$D_i g_i^z = \sin\theta_i \cos\theta_i \left(\frac{\chi_{i||}}{T_{1i}} - \frac{\chi_{i\perp}}{T_{2i}}\right)\{\omega_i \Gamma_i - (i\omega + \frac{1}{T_{2i}})\sin\theta_i \cos\theta_i \left(\frac{1}{T_{1i}} - \frac{1}{T_{2i}}\right)\} +$$

$$+ \overline{S_i^x}\{\omega_i \sin\theta_i \cos\theta_i \left(\frac{1}{T_{1i}} - \frac{1}{T_{2i}}\right) + \Gamma_i (i\omega + \frac{\sin^2\theta_i}{T_{1i}} + \frac{\cos^2\theta_i}{T_{2i}})\} +$$

$$+ \left(\frac{\cos^2\theta_i}{T_{1i}}\chi_{i||} + \frac{\sin^2\theta_i}{T_{2i}}\chi_{i\perp}\right) \times$$

$$\times \{\omega_i^2 + (i\omega + \frac{1}{T_{2i}})(i\omega + \frac{\sin^2\theta_i}{T_{1i}} + \frac{\cos^2\theta_i}{T_{2i}})\},$$

where

$$\mu_i^2 D_i = (i\omega + \frac{\sin^2\theta_i}{T_{1i}} + \frac{\cos^2\theta_i}{T_{2i}})(i\omega + \frac{1}{T_{2i}}) \times$$

$$\times (i\omega + \frac{\cos^2\theta_i}{T_{1i}} + \frac{\sin^2\theta_i}{T_{2i}}) +$$

$$+ 2\omega_i \Gamma_i \sin\theta_i \cos\theta_i \left(\frac{1}{T_{1i}} - \frac{1}{T_{2i}}\right) + \Gamma_i^2 (i\omega + \frac{\sin^2\theta_i}{T_{1i}} + \frac{\cos^2\theta_i}{T_{2i}}) +$$

$$+ \omega_i^2 (i\omega + \frac{\cos^2\theta_i}{T_{1i}} + \frac{\sin^2\theta_i}{T_{2i}}) -$$

$$- \sin^2\theta_i \cos^2\theta_i \left(\frac{i}{T_{1i}} - \frac{1}{T_{2i}}\right)(i\omega + \frac{1}{T_{2i}}).$$

Here we have used the abbreviations:

$$\sin\theta_i = \frac{\Gamma_i}{H_i}, \quad \cos\theta_i = \frac{\omega_i}{H_i}, \quad \omega_i = \mu_i \sum_j t_{ij} \mu_j \overline{S_j^z},$$

$$\chi_{i||} = \frac{1}{4kT}\left(1 - \tanh^2\left(\frac{H_i}{2kT}\right)\right),$$

and

$$\chi_{i\perp} = \tanh\left(\frac{H_i}{2kT}\right) / 2H_i .$$

References

1. *V.G.Vaks, N.E.Zein. FTT, 17, 1617 (1975).*
2. *B.Zeks. "Random Ferroelectrics" Preprint (1975).*
3. *K.Handrich. "Amorphe Magnetika" in Magn. Eigenschaften von Festrkörpern, VEB Deutscher Verlag Grundstoffindustrie, Leipzig (1975).*
4. *A.B.Harris "Magnetism in disordered systems" in ICM' 76 Proceedings Physica B (Febr. 1977).*
5. *R.Blinc, B.Zeks. Advances in Physics, 91, 693 (1972).*
6. *P.S.Peercy. Phys.Rev., B12, 2725 (1975).*
7. *This question was pointed out by Dr. R.Ferchmin in private communications.*
8. *S.Kirkpatrick. Rev.Mod.Phys., 45, 574 (1973).*
9. *A.B.Harris, P.L.Leath, R.J.Elliott, B.G.Nickel. J.Phys., C7, 1693 (1974).*
10. *K.D.Godzig, A.Blumen. phys.stat.sol.(b), 66, 569 (1974).*
11. *M.Tokunaga, T.Matsubara. Progr. theor. Phys. (Kyoto) 35, 581 (1966).*
12. *J.Holakovsky. Czechosl. Jour.Phys., B22, 651 (1972).*
13. *V.Kopshky. Czechosl.Jour.Phys., B21, 896 (1971).*
14. *R.Blinc, R.Pirc, B.Zeks. Phys.Rev., B13, 2943(1976).*
15. *H.Shiba. Progr. theor.Phys. (Kyoto), 46, 77 (1971).*
16. *F.Brouers, M.Cyrot, F.Cyrot-Lackmann. Phys.Rev., B7. 4370 (1973).*
17. *M.E.Lines. Phys.Rev., B9, 3927 (1974).*
18. *B.R.Cooper. Lecture at XI Annual Winter School for Theor. Phys. of the University of Wroclaw, Karpacz (1974).*
19. *F.Ducastelle. J.Phys., C4, 175 (1971).*
20. *H.Brüter, J.Kowalski. Physica, A, to be published.*
21. *C.Frenzel, B.Pietrass, E.Hegenbarth. phys.stat.sol. (a), 2, 273 (1970).*

Received by Publishing Department
on December 8, 1976.