

С 34272

S-81

СООБЩЕНИЯ
ОБЪЕДИНЕННОГО
ИНСТИТУТА
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ

Дубна

163/2-72

24/1-72

E4 - 6172

6172



ЛАБОРАТОРИЯ ТЕОРЕТИЧЕСКОЙ ФИЗИКИ

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**THEORY
OF COHERENT NEUTRON SCATTERING
BY HYDROGEN-BONDED FERROELECTRICS
AT LOW TEMPERATURES**

**II. Scattering Characteristics
and Various Conceptions
of the Tunnelling Quasi-Spin Model**

1971

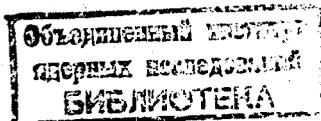
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S.Stamenković*

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**II. Scattering Characteristics
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of the Tunnelling Quasi-Spin Model**

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Стаменкович С.С.

E4-6172

Теория когерентного рассеяния нейтронов на сегнетоэлектриках
с водородной связью при низких температурах (II)

Для экспериментальной проверки закона дисперсии поляризационной ("квасиспиновой") моды колебаний рассматривается квазиупругое рассеяние нейтронов и рассеяние на малые углы в сегнетоэлектриках типа КДР. Обсуждаются различные концепции квазиспиновой модели с туннелированием, природа низкотемпературной полярной фазы, а также колебания туннелирующих частиц вдоль оси анизотропии в связи с когерентным рассеянием нейтронов.

Сообщения Объединенного института ядерных исследований
Дубна, 1971

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E4-6172

Theory of Coherent Neutron Scattering by
Hydrogen-Bonded Ferroelectrics at Low
Temperatures. II. Scattering Characteristics
and Various Conceptions of the Tunnelling
Quasi-Spin Model

To check on experimentally the dispersion law of the polarization ("quasi-spin wave-like") mode the characteristics of quasi-elastic neutron scattering at small angles are studied in KDP type ferroelectrics. The various conceptions of the tunnelling quasi-spin model, the nature of the low temperature polar phase as well as the vibration of tunnelling particles along the axis of anisotropy are discussed with reference to the coherent neutron scattering.

Communications of the Joint Institute for Nuclear Research.
Dubna, 1971

I. Introduction

In the preceding paper /1/ (referred to as I hereafter) we derived the general differential cross section for inelastic coherent neutron scattering by KDP type ferroelectrics. Using the tunnelling quasi-spin model and the model of two interacting harmonic oscillators as well as taking into account the proton (deuteron)-lattice interaction, this cross section was written as an explicit function of scattering momentum and energy transfers.

As pointed out by Villain and Stamenkovic /2/, in the coherent neutron experiments on a deuterated isomorphous sample ($KD_2 PO_4$), when the tunnelling frequency is very small, we have a chance of observing a weak \vec{q} -dependent collective frequency at $T < T_c$. However, De Gennes predicted that the scattering would be mostly elastic in this case /3/. Actually, it was found to be quasi-elastic as the range of the differential cross section is less than about 10^{11} Hz /4,5/.

On the basis of the previous approach, in this paper we proceed the study of the quasi-elastic neutron scattering characteristics in order to check on experimentally the dispersion law of the polarization ("quasi-spin wave-like") mode /2,6-10/. To do this we shall also consider the characteristics of the coherent neutron scattering at small angles where even for protons (in KH_2PO_4) one could expect considerable interference effects/11/. Finally, we try to interpret the existing variety of the tunneling quasi-spin model and to discuss the nature of the low temperature polar phase as well as the vibration of tunnelling particles (D or H) along the c -crystal axis with reference to the scattering properties of such ferroelectrics.

II. The Quasi-Elastic Coherent Scattering of Neutrons

In the study of coherent effects in KDP type ferroelectrics the general case of scattering geometry (incidence neutron beam, i.e. initial momentum of neutrons \vec{p} fixed by angles ϕ_0 and γ_0 with respect to the crystal axes a and c , respectively) is of great interest. As in this case the element of the solid angle is $d\Omega = \sin\gamma, d\gamma d\phi$, the scattering angle θ should be expressed in terms of spherical coordinates ϕ and γ of the outgoing neutron momentum \vec{p}' .

From the previous models (I), i.e. the estimates of the quasi-spin form factor and the Debye-Waller factor, one concludes that the differential cross section (I,(9)) is very anisotropic thought in such a simplified form.

This is predominantly expressed through the quasi-spin form factor as a function of the reciprocal lattice vector projected to the direction of a given deuteron bond

$$g_r = p (\sin \gamma \cos \phi - \sin \gamma_0 \cos \phi_0^r); \cos \phi_0^r = \begin{cases} \cos \phi_0, & \vec{l}_r \parallel \vec{a} \\ \cos(\pi/2 - \phi_0), & \vec{l}_r \parallel \vec{b} \end{cases} \quad (1)$$

On the other hand, at low temperatures the thermal vibrations of the lattice, taken into account by the factor $e^{-2W_q^r}$, should not affect essentially the intensity of the quasi-elastic coherent scattering.

In the preceding approximation of the real crystal structure by a Bravais lattice of $[K - PO_4]$ complexes with associated deuteron bonds (I), the index r indicates only different deuteron-bonding orientations (parallel (II) or perpendicular (I) to the a -axis), so that eight deuteron-like modes and their energy widths are truncated to one mode ω_q^- of the energy width Γ_q .

As the position of a coherent peak in the energy spectrum can be determined reliably enough, to verify the dispersion law obtained in the quasi-spin formalism it is sufficient to use the Dirac function instead of the expression for a Lorentzian line (I,9)^{x/}. After finding the form factor F_q^{\parallel} and F_q^{\perp} , according to the formula (I,(16) along with (29)), and the exponents of Debye-Waller factors W_q^{\parallel} and W_q^{\perp} , according to the formula (I,(17) along with (20), (24) and (25)), the differential cross section for coherent scattering of neutrons with emission (+) or absorption (-) of the quasi-spin wave becomes

^{x/}This is so much more valid since the polarization mode, although of very small energy, is of the optical type (see ref./12/) and because the elastic incoherent scattering can be subtracted quite accurately ^{74/}.

$$\frac{d^2 \sigma_{coh}}{d\Omega dE_p'} = \frac{Nn'}{2} \sigma_{coh} f_g \frac{p'}{p} \frac{H}{\omega_q} \left[n(\vec{q}) + \frac{1}{2} \pm \frac{1}{2} \right] \delta(E_p' - E_p \pm h\omega_q^-), \quad (2)$$

where

$$f_g = \left[|F_g^{\parallel}|^2 e^{-2W_g^{\parallel}} + |F_g^{\perp}|^2 e^{-2W_g^{\perp}} \right]. \quad (3)$$

III. The Scattering at Small Angles

Let us consider the characteristics of coherent scattering at small angles in more detail. With some modifications they will be similar to the corresponding properties of neutron scattering by ferromagnetics studied by Maleev /13/. From the energy conservation law

$$p'^2 = p^2 + \frac{2m_n}{h^2} [\omega_0 + \omega_1 (\vec{g} + \vec{q})^2], \quad (4)$$

for the outgoing neutron momentum one obtains a pair of values for each the scattering with emission (p'_{+}, p'_{-}) and with absorption (p'_{+}, p'_{-})

$$p'_{+,-} = \frac{B|\vec{P}|}{B+1} \left\{ \cos \theta \pm \sqrt{\cos^2 \theta - \cos^2 \theta_0^+} \right\} \quad (5)$$

$$p'_{+,-} = \frac{B|\vec{P}|}{B-1} \left\{ \cos \theta \pm \sqrt{\cos^2 \theta - \cos^2 \theta_0^-} \right\}, \quad (6)$$

Here θ is the scattering angle with respect to the direction of the vector $\vec{P} = \vec{p} + \vec{g}$ and

$$\cos^2 \theta_0^\pm = \frac{B \pm 1}{B} \left[1 - \frac{p^2 + A}{B P^2} \right]; \quad A = 2m_n \omega_0 / h^2, \quad (7)$$

$$B = 2m_n \omega_0 / h^2.$$

After integration in energies of scattered neutrons for the angular distribution one obtains ^{x/}

$$\frac{d\sigma_{coh}^\pm}{d\Omega} = N n' \sigma_{coh} f_g \frac{PB}{P(B \pm 1)^2} (\cos^2 \theta - \cos^2 \theta_0^\pm)^{-1/2} \left[[n(q_\pm^+) + \frac{1}{2} \pm \frac{1}{2}] \right]. \quad (8)$$

$$(\cos \theta + \sqrt{\cos^2 \theta - \cos^2 \theta_0^\pm})^2 + [n(q_\pm^+) + \frac{1}{2} \pm \frac{1}{2}] (\cos \theta - \sqrt{\cos^2 \theta - \cos^2 \theta_0^\pm})^2 \},$$

$$q_+^\pm = [P^2 B / (B \pm 1)^2] \left[\pm (\cos \theta + \sqrt{\cos^2 \theta - \cos^2 \theta_0^\pm})^2 \pm ((B \pm 1)/B)^2 \frac{p^2}{P^2} \right], \quad (9)$$

$$q_-^\pm = [P^2 B / (B \pm 1)] \left[\pm (\cos \theta - \sqrt{\cos^2 \theta - \cos^2 \theta_0^\pm})^2 \pm ((B \pm 1)/B)^2 \frac{p^2}{P^2} \right]. \quad (10)$$

As it can be seen the scattering depends essentially on the parameter B . The preliminary estimates give that $B \approx 100$. From the definition of the angle θ_0^- (7) it follows that

$$\cos^2 \theta_0^- > 1 - 1/B, \quad (11)$$

^{x/}The contribution of Jacobian $J = \left| 1 \pm \frac{h}{2E_p} \vec{p}' \cdot \nabla_{\vec{q}} \omega_{\vec{q}}^- \right|$ is neglected here and later.

respectively

$$\theta_0^- < B^{-1/2} \approx 1/10. \quad (12)$$

Since

$$1 > \cos \theta \geq \cos \theta_0^- > 0, \quad (13)$$

then

$$\theta < B^{-1/2}. \quad (14)$$

This means that the scattering with absorption is possible only in the narrow conus with the axis $\vec{P} = \vec{p} + \vec{g}$, provided that two values of scattering momentum of neutrons q_+ (9) and q_- (10) /14/ correspond to every scattering direction in this conus. As $\cos^2 \theta_0^- < 1$ one obtains that the scattering with absorption is possible only if the following condition is fulfilled

$$\cos \eta > \frac{1}{2} \left\{ \frac{A}{pg} \frac{B-1}{B} - \frac{g}{p} - \frac{p}{Bg} \right\}, \quad (15)$$

where η is the angle between \vec{P} and \vec{g} , i.e. for

$$p \geq Bg \left(\sqrt{1-1/B + A(B-1)/B^2 g^2} - 1 \right). \quad (16)$$

In case of the scattering with emission of the quasi-spin wave analogous conditions can be obtained

$$\cos \eta < \frac{1}{2} \left\{ \frac{A}{pg} \frac{B+1}{B} - \frac{g}{p} + \frac{p}{Bg} \right\}, \quad (17)$$

respectively

$$p \geq Bg \sqrt{1 + 1/B - A(B+1)/B^2 g^2 - 1}. \quad (18)$$

Here two cases should be considered:

$$a) \quad [1 - (p^2 + A)/BP^2] > 0; \quad (19)$$

$$b) \quad [1 - (p^2 + A)/BP^2] < 0. \quad (20)$$

For the case a), as in the scattering with absorption, $\cos \theta$ changes in the same, i.e. analogous interval $(\theta_0^- \rightarrow \theta_0^+)$ (13) and two values of scattering neutron momentum q_+^+ (9) and q_-^+ (10) correspond to every scattering angle.

For the case b), the scattering is possible only if the following condition is fulfilled

$$[Bg - \sqrt{Bg^2 + A(B-1)}] / (B-1) < p < [Bg + \sqrt{Bg^2 + A(B-1)}] / (B-1). \quad (21)$$

From (5) and (7) it follows that $p_-^+ < 0$, i.e. that in this case only one value of the scattering neutron momentum p_+^+ corresponds to every scattering angle. Here the scattering would also be possible at considerably greater angles as, theoretically, the scattering angle can vary from 0 to π .

Finally, from the conditions (15) and (17) it follows that the simultaneous scattering with absorption and emission of the quasi-spin wave is possible if

$$\left[\frac{A}{pg} \frac{B-1}{B} - \frac{g}{p} - \frac{p}{Bg} \right] \leq 2 \cos \eta \leq \left[\frac{A}{pg} \frac{B+1}{B} - \frac{g}{p} + \frac{p}{Bg} \right], \quad (22)$$

i.e. in the region of values η where both cross sections reach the maximum values. For this simultaneous scattering the whole differential cross section can be obtained from (I, (9))

$$\frac{d^2 \sigma_{coh}}{d\Omega dE_p'} = 2 N n' \sigma_{coh} \frac{p'}{p} f_g \frac{H}{\omega_q^-} \frac{[n(\omega_q^-) + 1] \Gamma_q E^2}{[E^2 - (h\omega_q^-)^2]^2 + (2E\Gamma_q)^2}, \quad (23)$$

where $E = |E_p - E_p'|$.

If one presents the complex polarization mode $(h\omega_q^- + i\Gamma_q)$ in the form $\{ [E_q^2 - (\gamma_q/2)^2]^{1/2} + i(\gamma_q/2) \}$ and according to $E \approx h\omega_q^- \approx E_q$, the whole differential cross section can be written in the form of Scalyo et al.^{/15/x/}, where the "quasi-spin-Debye-Waller factor" (3) stands in - stead of the phonon inelastic structure factor:

$$\frac{d^2 \sigma_{coh}}{d\Omega dE_p'} = \frac{N n'}{\pi} \sigma_{coh} \frac{p'}{p} f_g H [n(E) + 1] \frac{E \gamma_q}{(E^2 - E_q^2)^2 + (E \gamma_q)^2}. \quad (24)$$

The quasi-elastic energy width γ_q' of the differential cross section $d\sigma_{coh} / d\Omega$ can be estimated after integrating the expression (24) in transfer-energies. For the sake of simplicity one can take that $\vec{q} = 0$, and then obtains

^{x/}Their effective cross section refers to the paraelectric phase, provided that the boson distribution is substituted by Boltzmann's. In the general form the expression for the cross section remains the same for $T < T_c$ as well.

$$\frac{d\sigma_{coh}}{d\Omega} \approx Nn' \sigma_{coh} f_g H \sqrt{\frac{|E_p + E|}{E_p}} \left[1 + e^{-\frac{E_0}{k_B T}} \right] \frac{1}{2E_0}, \quad (25)$$

with a quasi-elastic energy broadening $\gamma'_0 = \pi \gamma_0$.

IV. Discussion of Scattering Characteristics and Various Conceptions of the Tunnelling Quasi-Spin Model

The differential cross section $\frac{d^2 \sigma_{coh}}{d\Omega dE_p}$, i.e. $\frac{d\sigma_{coh}}{d\Omega}$ for inelastic coherent neutron scattering is related to a particular point in the reciprocal lattice space \vec{q} . Actually, this scattering can be observed only if f_g is not small. Thus, if \vec{q} , i.e. \vec{q} , is direct along c -axis, the scattering intensity is very small being proportional to the overlapping integral $\int \psi_L \psi_R d\vec{R} = e^{-(\ell/2b)^2}$ (which is practically equal to zero) and to the Debye-Waller factor reduced to a minimum (also very small) value. Using the obtained expressions for the differential cross sections one can check on experimentally very interesting characteristics of the neutron scattering as expected by the present theory. For instance, if \vec{q} is in the plane a - b and perpendicular to one half of deuteron bonds (they lie almost parallel to either a or b directions) then one obtains approximately the intensity of neutrons scattered on the other half of deuteron bonds parallel to scattering momentum transfer. A similar situation appears when \vec{q} is in one of the two planes normal to the deuteron-bond direction.

If $\vec{g} = 0$, then it corresponds to the scattering at small angles. In this case, owing to f_g ($\vec{g} = 0$), the scattering intensity will also be small. However, the considered scattering characteristics can again be observed very well by the monochromatic neutron beam. The advantage of the scattering at small angles lies in the possibility to carry out experiments with the polycrystal. Besides, the scattering by acoustic phonons is small (or absent completely if $v_n < v_z = v_{ac}$) and the whole scattering should be mostly originated by the polarization mode.

If $\vec{g} \neq 0$, by changing the orientation of the monocrystal one can select the scattered neutrons of fixed energy (the scattering at small angles is now taken with respect to \vec{P}). Hence, one can immediately draw out some conclusions concerning the dispersion law of the polarization wave. Namely, in the dispersion law for small \vec{q} there is a constant and quadratic term (at low temperatures the polarization mode is approximately of the form $\omega_q = \omega_0 + \omega_1 q^2$ /10/). As a consequence one should expect that the scattering intensity becomes relatively large for small inclinations from the Bragg angle. It is known that in the linear dispersion law there is no similar increase of this intensity. This means that at small angles one should expect the main contribution from the polarization mode itself /16/.

Besides the already analysed scattering characteristics at small angles let us mention that by such a scattering the parameters of the approximative dispersion law ω_0 and ω_1 can be directly determined. These terms can be connected with the experimentally observable quantities in the simplest possible way if the formulae (5) and (6) are used.

From the difference of the two possible values for the scattering neutron momentum directed to the centre of the inelastic peak ($\theta = 0$), from (5) and (6) one obtains

$$[p'_+ - p'_-]_{\theta=0} = \frac{2BP}{B-1} \sin \theta_0^-; \quad [p'_+ - p'_-]_{\theta=0} = \frac{2BP}{B+1} \sin \theta_0^+. \quad (26)$$

Since θ_0^- and θ_0^+ depend on the parameters A and B , ω_0 and ω_1 can be directly determined as well as their changes with temperature or upon the applied electrical field.

Experimental verification of the considered characteristics of the coherent neutron scattering has even more pronounced significance. Namely, experimental fitting of the energy parameters in accordance with their theoretical estimates could point at the proper understanding of the quasi-spin model with respect to the existing variety of its interpretations.

As it is known, recent experiments of incoherent neutron scattering /17,18/ have shown good agreement with the simple "tunnel-scattering" model of Stiller and Stamenkovic /19,20/. Reproducing the previous results of Bacon and Pease /21/ and confirming the existence of an interference effect within a single hydrogen bond /19,20/, these experiments proved that protons (deuterons) really fluctuate between two equilibrium positions. However, the tunnelling itself can be introduced in the quasi-spin model for the ferroelectric crystal as a whole either in accordance with the very well known rules of Slater /22/ (see also ref. /23/) or according to Blic /24/, De Gennes /3/ and Matsubara and Tokunaga /6/, whose Hamiltonian does not

allow the reversal of a single H -bond. Since the light scattering has been observed for $T > T_c$. /25/, Villian and Aubry /23/ conclude that KDP is far enough from Slater's limit although the mechanism of "closed loops" can also be presented but with a characteristic frequency likely smaller than in Blic's mechanism. On the other hand, the theoretical conceptions of the Blic model have recently been essentially reconsidered by Novakovic /8/. Using the second quantization formulation different than in ref. /6/, this author never referred to any kind of "tunnelling" so long as a system of a large number of particles is considered. By such an approach, independent of Stasyuk's idea /9/, the complete set of elementary excitations (of the Frenkel-exciton type) was approximated with the first two (ϵ_0 and ϵ_1) and reduced to the quasi-spin Hamiltonian for temperature close to the phase transition point. Therein the energy difference at a given hydrogen-bond Bravais lattice site ($\epsilon_1 - \epsilon_0$) was interpreted as a change of the kinetic energy when the proton makes a transition from $s_z = 1/2$ to $s_z = -1/2$. As a remarkable result it was shown that the energies of the proton configurations (the Slater-Takagi, i.e. the Blic parameters ϵ , w and w_1) are not mutually independent when one looks at the crystal as a whole. These parameters were also related to the parameters entering in the quasi-spin Hamiltonian. If one keeps in mind that the quasi-spin Hamiltonian makes sense for any temperature if 2Ω and J are temperature dependent (irrespective of some troubles concerning definition of the ground state /26/) as well as fair agreement between theory /8/ and experiments of Kaminow et al. /25,27/ and Buyers et al. /4/, then the experiments of neutron scattering at low temperatures and at small angles could have fundamental meaning for the

proper interpretation of the quasi-spin model. Actually, independent of the two possible tunnelling mechanisms, $2Q$ is very small at low temperatures and its direct measurement is not an easy task /28/. In any case the measurements in the temperature region of T_0 (a temperature characterizing a quasi-equilibrium state between subsystems of deuteron and phonon excitations /10/) would be of particular interest as for the maintenance of Slater's rules around this temperature one should expect to hold true the condition $T_0 > T_c$ (a transition temperature from total to partial ordering /23,28/). Of course, the neutron experiments at small angles can be performed in the vicinity of T_c as well, or for $T < T_c$ (or $T_c/2 < T < T_c$). In that case, the scattering characteristics due to mere specificity of the dispersion law of the ferroelectric mode would be considerably different and, judging by all things, one should rather relate them directly to the dielectric properties of the crystal. Besides other troubles concerning the consistency between theory and experiments /4,15,29/ let us only mention that at these temperatures arise the difficulties connected with the appearance of acoustic vibrations, the small intensity of considerably damped ferroelectric mode as well as with the presence of dipole interactions and kinematical effects (these latter coming from an exact boson representation /30/ of the quasi-spins (see ref. /10/)). In order to obtain a reasonable scattering intensity the process in which the neutrons give energy to the collective excitations should be investigated. In this respect the method of Zivanovic et al. /31/ as well as that of Stiller and coworkers /32,18/, if applied to test here considered scattering characteristics, seems to give exhaustive results.

A word would be added here as a general comment. The quasi-elastic nature of neutron scattering by KD_2PO_4 indicates that the low temperature polar phase (as the transition point /4/ as well) is determined by equilibrium between deuteron and phonon excitations (studied in an earlier paper /10/) rather than by the dynamical properties of this ferroelectric.

In the conclusion we shall briefly discuss the deuteron vibrations along the axis of anisotropy (identical with c -axis). The neutron diffraction experiments of Scalzo et al. /15/ have shown that the process of deuteration has a more pronounced effect. A striking result was the large distortional movement in the x - y plane of the oxygen tetrahedra as well as a rather large motion of the deuterium atoms along c -axis in phase with the P atoms (not in phase with the K atoms, as suggested by structural change at the transition point in KH_2PO_4 /21/). To compare obtained experimental results with theory, Scalzo et al. used the angular cross section according to the standard procedure for the neutron scattering by phonon excitations. However, the potential field of a deuteron is so anharmonic that the elementary excitations can by no means be described as phonons although the ferroelectric mode is explicitly introduced in the cross section, i.e. in /15/.

As it is known, from Shur's irreducible representations /33/ Γ_3 ($T > T_c$) and Γ_1 ($T < T_c$), the linear combinations of 7, i.e. 13 basic vectors, associated with possible atomic displacements within a primitive cell, allow the D_z -vibrations as well in distinction from a dynamical model of Cochran /29,34/, Villain-Stamenkovic /2/ and Kobayashi /7/ (the latter authors use the tunnelling

quasi-spin model extended to the optical mode of the K and P ions along the c -axis) which admits only D_x (or D_y) motion. We shall restrict ourselves only to the D_z -vibrations as O -vibrations can in principle be accounted by the Debye-Waller factor (I, (17)). In the quasi-spin formalism the D_z -vibrations can be accounted for additionally by means of the correction of hybridized optical branches ω_q^- and ω_q^+ - depending on whether D_z -vibrations are in phase with the K or P atoms.

It is less probable that interactions of heavy ions and quasi-spins with real spins of tunnelling particles (D or H) have some influence on the correlation in atomic motions. Otherwise, considering the smallness of the deuteron tunnelling at the ground state /8/ it is not excluded that $2\Omega_D$ is truly negligible as compared to the energy splitting of the first excited state. So the corresponding triggering mechanism could be associated with the next pair of localized wave functions, $\Psi_{L,R}^1 = \Psi_s^1 + \Psi_o^1$ (Ψ_s^1, Ψ_o^1 - the symmetrical eigenfunctions of the double degenerated first excited state). As a consequence, it might appear that the interaction between deuterons and heavy ions-via these first excited states results in D_z -vibrations in-phase with P atoms. This mechanism is already included in the templet of Novakovic ($2\Omega_D^1 = \epsilon_2 - \epsilon_1 \gg 2\Omega_D = \epsilon_1 - \epsilon_0$; $J_{II} \rightarrow J_{II}^{(1,1)} = A_n(1,1)$ /8/, or $J_{II}^{(1,1)} = V_{II}(\Psi_{L1}^1, \Psi_{L1}^1$;

$\Psi_{L1}^1, \Psi_{L1}^1)$ /6/) thus favouring it once more. Anyhow, "the problem of phases", being of great theoretical interest, remains open as yet, but it would be worthwhile to perform the parallel measurements on KD_2PO_4 and KH_2PO_4 , especially at low temperatures.

Concluding this paper we notice that a more concrete calculation based on the theory presented in this paper is going on.

Acknowledgement

The author would like to acknowledge Dr.L. Novakovic for very useful discussions on the subject.

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Received by Publishing Department
on December 15, 1971.