

ON THE TUNNELLING CONCEPT IN THE GENERAL THEORY OF
FERROELECTRIC PHASE TRANSITIONS

S. Stamenković (a), N.M. Plakida (b), V.L. Aksienov (b)
and T. Siklos (c)

(a) Institute "Boris Kidrič"-Vinča, Beograd, Yugoslavia

(b) Joint Institute for Nuclear Research, Dubna, USSR

(c) Central Research Institute for Physics, Budapest, Hungary

Abstract

The unified tunnelling model Hamiltonian describing both "order-disorder" and "displacive" ferroelectric phase transitions is proposed by introducing the non-orthogonal (overlapping) "eigenbasis" in the pseudospin space.

In the last several years a considerable effort has been made to describe the ferroelectric phase transitions of various types ("order-disorder", "displacive" and mixed) in the frame of only one universal model¹⁻⁶. In the meantime we also have tried to formulate the general ferroelectric model⁷ by introducing two order parameters, the average population ($\sigma_\alpha(T) = \frac{1}{2}(1 + \alpha\sigma_2)$) for one of two equilibrium positions, $\alpha = +, -$, and the average displacement ($\eta_\alpha(T) = \sqrt{\frac{B}{A}} b_\alpha(T)$) of active atoms as determined in the scheme of the self-consistent pseudospin-phonon approach (see, also, Ref. 8).

However, so far the inherent quantum-mechanical effect manifested in an occasional jumping from one well to another (in a single-particle double minimum potential) has not been explicitly taken into account. As an exact analytic description of whole dynamics is rather unfeasible⁹, we propose the "left-right" representation¹⁰ of our previous Hamiltonian⁷ in the non-orthogonal pseudospin "eigenbasis". In such a way, in addition to the conserved phonon picture (which is missing in earlier approaches^{11,12}) the statistical disorder and tunnelling motion of active atoms are also incorporated as an additional degree of freedom. Further, having approximated, respectively, the single-particle double well potential by an anharmonic oscillator potential and atomic pair-interaction only by harmonic terms, the hybridized tunnelling-pseudospin-phonon Hamiltonian we need becomes*.

$$\begin{aligned}
 H = & \frac{1}{2} \sum_{i\alpha} \left(\frac{1}{2m} \vec{p}_{i\alpha}^2 - \frac{A}{2} \vec{s}_{i\alpha}^2 + \frac{B}{4} \vec{s}_{i\alpha}^4 \right) (1 + \epsilon \sigma_{ix} + \alpha \sqrt{1 - \epsilon^2} \sigma_{iz}) + \\
 & \frac{1}{4} \sum_{\substack{i=1 \\ \alpha, \beta}} \phi_{ij}^n (\vec{s}_{i\alpha} - \vec{s}_{j\beta})^2 \left(1 + \frac{\epsilon}{2} \sigma_{ix} + \alpha \frac{\sqrt{1 - \epsilon^2}}{2} \sigma_{iz} + \right. \\
 & \left. \frac{\epsilon^2}{4} \sigma_{ix} \sigma_{jx} - \frac{\sqrt{1 - \epsilon^2}}{2} \sigma_{ix} \sigma_{jz} - \alpha \beta \frac{1 - \epsilon^2}{4} \sigma_{iz} \sigma_{jz} \right).
 \end{aligned}$$

Here $\vec{R}_{i\alpha} = \vec{r}_i + \sigma_i^{+++} \vec{s}_i + \sigma_i^{-+} \vec{s}_i$ and $\vec{p}_{i\alpha} = m \dot{\vec{s}}_{i\alpha}$ ($i, j = 1, 2, \dots, N$) are the coordinate and momentum of active atom $\vec{s}_{i\alpha} = \vec{b}_{i\alpha} + \vec{u}_{i\alpha}$

*Of course, the physical concept herein is essentially different from the pseudospin-phonon model in KDP-crystals¹³.

being the sum of its off-center equilibrium displacement ($\vec{b}_{1\alpha}$) and the thermal fluctuation ($\vec{u}_{1\alpha}$); σ_{ix} and σ_{iz} are x and z components of the pseudospin operator; A and B define, respectively, the height of the potential barrier, $U_0 = A^2/(4B)$, and the distance between the two potential minima, $d_0 = 2\sqrt{A/B}$ whereas ϕ_{ij}'' is the harmonic coupling constant; The overlapping constant ϵ is estimated in some model calculations (see, for instance, Refs. 14-18).

As to conclude we hope that our novel unified model could give some initial hint to yield deeper insight into the nature of ferroelectricity at all.

References: 1-6. The same from 1-6. in Ref.7; 7. S.Stamenković, N.M.Plakida, V.L.Aksienov, T.Siklos, Ferroelectrics 14,655,1976; Phys.Rev./B/, december 1976; 8. N.M. Plakida, Phys. Lett. 32A,134,1970; 9. H.Beck, J.Phys. C: Solid St.Phys. 9,33,1976; 10. P.G.De Gennes, Solid State Commun. 1,132,1963; 11. M.Tokunaga, T.Matsubara, Progress Theor.Phys. 35,581,1966. 12. V.G. Vaks, Introduction to the Microscopic Theory of Ferroelectrics, Chap. 5, ed. "Nauka", Moscow 1973 /in Russian/; 13. J.Villain, S.Stamenković, phys. stat. sol. 15,585,1966; 14. S.Stamenković, FTT 10,861,1968 /in Russian/; 15. J.B. Coon, N.W. Naugle, R.D. McKenzie, Journ. Spectr. 20,107,1966; 16. E.Merzbacher, Quantum Mechanics, Chap.5, John Wiley, New York 1970. 17. E.A.Pschenichnov, N.D.Sokolov, Opt. Spectr. 11,16,1961; 17,343,1964 /in Russian/; 18. G.Biczko, J.Ladic, Acta Phys. Hung. 20,11,1966.