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ABSTRACT

The unified model Hamiltonian describing both "order-disorder" and "displacive" type ferroelectrics is extended by introducing simultaneously right from the beginning the phonon vibrations, statistical disorder and tunnelling motion of active atoms.

АННОТАЦИЯ

Модельный гамильтониан, описывающий сегнетоэлектрические фазовые переходы как типа "порядок-беспорядок", так и типа "смещения"обобщается для одновременного учета всех типов движения активных атомов: фононных вибраций, статистического упорядочения и эффекта туннелирования.

KIVONAT

A "rend-rendezetlen" tipusu és a "rácstorzulással járó" ferroelektromos fázisátalakulásokat egyaránt jól leiró modell Hamilton-operátort ugy általánositjuk, hogy az az aktiv atomok mindhárom tipusu mozgásának - vagyis a hőrezgéseknek, a statisztikai rendeződési folyamatokkal, valamint az alagut-effektussal kapcsolatos mozgásoknak - szimultán figyelembevételére alkalmas legyen.

The authors have recently proposed the unified theory of ferroelectrics [1] taking into account both statistical disorder and phonon vibrations of active atoms. Starting from quite a general Hamiltonian

$$H = \sum_i \left\{ \frac{\vec{p}_i^{\,2}}{2m} + V(\vec{R}_i) \right\} + \frac{1}{2} \sum_{i \neq j} \varphi(\vec{R}_i - \vec{R}_j), \qquad /1/$$

by representing the atomic coordinate as

$$\vec{R}_i = \vec{l}_i + \sigma_i^+ \vec{S}_i^+ + \sigma_i^- \vec{S}_i^- \qquad /2/$$

and by expressing the potential energy by the first two terms of the Taylor expansion, some kind of a hybridized phonon-Ising Hamiltonian has been obtained

$$H = \sum_{i,\alpha} \sigma_{i\alpha} \left\{ \frac{\vec{p}_{i\alpha}^{\,2}}{2m} - \frac{A}{2} \vec{S}_{i\alpha}^{\,2} + \frac{B}{4} \vec{S}_{i\alpha}^{\,4} \right\} + \frac{1}{4} \sum_{\substack{i \neq j \\ \alpha, \beta}} \sigma_{i\alpha} \sigma_{j\beta} \varphi_{ij}'' (\vec{S}_{i\alpha} - \vec{S}_{j\beta})^2. \qquad /3/$$

$$(\alpha, \beta = +, -)$$

In the above expressions $\vec{l}_i$ denotes the atomic site-position; $\vec{S}_{i\alpha} = \vec{b}_{i\alpha} + \vec{u}_{i\alpha}$ is the sum of the off-center equilibrium displacement $(\vec{b}_{i\alpha})$ and the thermal fluctuation $(\vec{u}_{i\alpha})$ of the atom moving in a double well potential $V(\vec{R}_i)$; $\vec{p}_{i\alpha} = m \dot{\vec{S}}_{i\alpha}$ represents the "left" $(+)$ and "right" $(-)$ atomic momentum; $\sigma_{i\alpha} = \frac{1}{2}(1 + \alpha \sigma_{iz})$ is the projection operator simply expressed by the z-pseudospin component; $\varphi(\vec{R}_i - \vec{R}_j)$ is the familiar interaction between a pair of atoms $(i, j = 1, 2, \ldots, N)$; A and B define, respectively, the height of the potential barrier, $U_0 = A^2/4B$, and the distance between the two potential minima, $d_0 = 2\sqrt{A/B}$.

After the self-consistent pseudospin-phonon procedure a closed system of equations for two order parameters $(\sqrt{B/A}\langle b_{i\alpha}\rangle = \eta_\alpha$ and $\langle \sigma_{iz}\rangle = \sigma_z)$ as functions of the reduced coupling constant $(f_0 = \frac{1}{A}\sum_i \varphi''_{ij})$ and temperature has been obtained and solved numerically. Further it follows that the ferroelectric phase transition can be either of the order-disorder type or of the displacive type or mixed; (in these cases the transition of I, II or mixed order is possible) - depending on the ratio of two--particle potential to the single-particle potential. However, in this model the inherent quantum-mechanical effect, manifested in a single-particle tunnelling motion of active atoms between the two potential minima has not been explicitly taken into account.

Such a peculiar motion imposes some kind of the over-Heisenberg uncertainity relation (in the coordinate or pseudospin picture)

$$\langle(\Delta\hat{p})^2\rangle\langle(\Delta\hat{R})^2\rangle \simeq \left\langle\left(\frac{2\Omega m d_0}{\hbar}\sigma_y\right)^2\right\rangle\left\langle\left(\frac{d_0\sigma_z}{2}\right)^2\right\rangle \gg \frac{\hbar^2}{4}, \qquad /4/$$

i.e.,

$$d_0\sqrt{2\Omega m} \gg \hbar, \qquad /5/$$

(m - the mass of active atom and $2\Omega \simeq \int \Psi_l(R_i) H_i \Psi_r(R_i) dR_i$; $\Psi_l(R_i)$ and $\Psi_r(R_i)$ being, respectively, the real wave functions of the "left" and "right" atomic states of the single-particle Hamiltonian, H_i ; see, also, Refs. [2], [3])[x] for which no satisfactory "narrowing" has been obtained so far, even theoretically.

As an exact analytic description of dynamics induced by /1/ or /3/ is rather impossible we propose a novel approximative scheme which takes into account all the intriguing features of the ferroelectric phenomenon, i.e., the tunnelling, statistical disorder and phonon vibrations of active atoms, in the frame of only one universal model.

To realize such a concept we introduce the "left-right" representation of the Hamiltonian /3/ using the non-ortogonal pseudospin "basis"

$$|l\rangle = a\binom{1}{0} + b\binom{0}{1}\ ; \quad |r\rangle = a\binom{0}{1} + b\binom{1}{0}\ ;$$

$$(0 < a \lesssim 1 ;\ b \ll a), \qquad /6/$$

which corresponds to the atomic localization in the left (l) or right (r) single-particle potential well in the limit of a non-penetrating barrier $(U_0 \to \infty)$. The coefficients a and b are determined from the completness condition $(a^2 + b^2 = 1)$ jointly with the overlap integral $(\varepsilon = 2ab = \int \Psi_l(R_i)\Psi_r(R_i)\, dR_i)$.

After a straightforward procedure the Hamiltonian /3/ is cast in the form we need

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x/ To explicit these functions, some approximative calculations can be used [4] - [7] provided that the ground state doublet yields a predominant contribution.

$$H = \frac{1}{2} \sum_{i\alpha} \left\{ \frac{\vec{p}_{i\alpha}^{\,2}}{2m} - \frac{A}{2} \vec{S}_{i\alpha}^{\,2} + \frac{B}{4} \vec{S}_{i\alpha}^{\,4} \right\} \left\{ 1 + \varepsilon \sigma_{ix} + \alpha \sqrt{1-\varepsilon^2}\, \sigma_{iz} \right\} +$$
$$+ \frac{1}{4} \sum_{\substack{i \neq j \\ \alpha, \beta}} \varphi_{ij}'' \left(\vec{S}_{i\alpha} - \vec{S}_{j\beta} \right)^2 \left\{ 1 + \frac{\varepsilon}{2} \sigma_{ix} + \alpha \frac{\sqrt{1-\varepsilon^2}}{2} \sigma_{iz} + \right.$$
$$\left. + \frac{\varepsilon^2}{4} \sigma_{ix} \sigma_{jx} + \frac{\sqrt{1-\varepsilon^2}}{2} \sigma_{ix} \sigma_{jz} + \alpha\beta \, \frac{1-\varepsilon^2}{4} \sigma_{iz} \sigma_{jz} \right). \qquad /7/$$

It is evident that this Hamiltonian takes the previously cited form /3/ for $\varepsilon = 0$.

Advantages of such a novel model Hamiltonian are obvious: in addition to the conserved phonon picture (which is missing in earlier approaches [3], [8]) the tunnelling motion is also incorporated as an additional degree of freedom (the displacements and the pseudospins are mutually independent but time-dependent variables). Therefore, our new model can reproduce the pseudospin (diffusive or polarization) waves abreast to all phonon modes in the system as well as the collective motion of atomic clusters.

In the conclusion we point out that the present model is favoured by recent computer simulations [9] and, among others [10], should be invoked for reasons of universality (see [10] and cross references). Finally, the model proposed here by means of a more accurate self-consistent procedure (including the symmetry point of view at zero temperatures [11], [12]), could give an initial hint for deeper insight into the nature of ferroelectricity in general. Such investigation in addition to the previous analysis, will be the subject of our future work.

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