THERMODYNAMICS OF QUANTUM LATTICE SYSTEM WITH LOCAL MULTI-WELL POTENTIALS: DIPOLE ORDERING AND STRAIN EFFECTS IN MODIFIED BLUME–EMERY–GRIFFITHS MODEL

Thermodynamics of quantum lattice system with local multi-well potentials: dipole ordering and strain effects in modified Blume–Emery–Griffiths model

Abstract. Some aspects of thermodynamics of the quantum lattice model with the local anharmonic potentials are considered for the case of deformed lattice. The effects, taking place under external pressure, are investigated in the framework of the Blume–Emery–Griffiths model corresponding to the local potential with three minima. The deformable BEG model is proposed for this purpose. Based on this model, the pressure dependences of the \( u = \frac{\Delta V}{V} \) volume deformation are calculated on an example of the \( \text{Sn}_2\text{P}_2\text{S}_6 \) crystal. The presence of anomalies of \( u(p) \) function in the regions of ferroelectric phase transitions as well as the tricritical point is established; the behaviour of the volume compressibility in these cases is investigated.

Подається в Phase Transitions
Submitted to Phase Transitions

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Institute for Condensed Matter Physics 2018
1. Introduction

Lattice models play an important role as one of extensions of statistical physics on new objects and phenomena in the field of condensed matter physics (especially, the physics of solid state). Relatively simple quantum lattice models often demonstrate a variety of phase states and sophisticated phase diagrams. As a widely used example, one can mention the description of thermodynamics and study of the order–disorder phase transitions in the crystals with locally anharmonic structure elements by the quantum lattice models. Local anharmonic potentials with multimimima shapes commonly occur in this case. The various localizations of particles correspond to different quantum states (configurations of structure elements). For systems with two-well local potentials (e.g. crystals with hydrogen bonds) this approach leads to the transverse Ising model (known in the theory of ferroelectrics as the de Gennes model [1]. In the case of the three-well symmetrical potential and at the same conditions an appropriate lattice model corresponds to the Blume–Emery–Griffiths model [2]. The model can be applied to description of crystals belonging to the Sn$_2$P$_2$S$_6$ family (with the possible partial substitutions Sn → Pb and S → Se, see [3]) which are an example of such objects.

The Blume–Emery–Griffiths model in its initial (pseudospin) formulation is a generalization of the $\mathbb{S} = 1$ Ising model. Alongside with the bilinear $(J)$ and biquadratic $(V)$ pair interactions, the model takes into account the single-ion anisotropy $(\Delta)$ describing the energy gap between states with $S^z_i = \pm 1$ and $S^z_i = 0$. A wide area of its application extends from phase transitions in $^3$He–$^4$He mixtures [2] to binary alloys and magnetic materials [4–11]. A special feature of the model manifests in the possible switch of the phase transition order from the second to the first one (a transition between phases with $\langle S^z \rangle = 0$ and $\langle S^z \rangle \neq 0$) and an appearance of the respective tricritical point (depending on ratios $J/\Delta$ and $V/J$ of model parameters).

From this point of view, the application of the BEG model to the Sn$_2$P$_2$S$_6$ crystal proved to be perspective. Primarily, the performed ab initio calculations [6] showed that ionic groups P$_2$S$_6$ exist in three configurations (determined by their form and distribution of electronic charge), which in the paraelectric phase are described by a symmetrical three-well potential in the energy space. In the absence of external influence, the Sn$_2$P$_2$S$_6$ crystal exhibits the second order phase transition to the ferroelectric phase at $T_c = 337$ K due to a dipole ordering of the aforementioned structure elements [7].

Secondarily, the thermodynamics of the Sn$_2$P$_2$S$_6$ crystal is sensitive
to the external hydrostatic pressure and to the partial substitutions Sn → Pb and S → Se. For example, at the increase of pressure the temperature $T_c$ of the second order phase transition decreases, a tricritical point is achieved at $T_{TCP} = 220$ K and, finally, the ferroelectric state is suppressed at $p = p^* = 1.5$ GPa. This effect can be explained by the influence of the pressure on the $\Delta$ parameter value as it was done in paper [3].

At the same time, such an effect needs a more detailed treatment. From the microscopic point of view, an immediate factor of the pressure influence is the induced deformation of the lattice. Respective changes in the environment of the local anharmonic structure elements (groups $P_2S_6$) affect the energy balance between various configurations. This is a source of the interaction between the deformation and local energy states (configurations of ionic groups). Such an interaction can be taken into account in the framework of a more complete description of the strain phenomena in the Sn$_2$P$_2$Se$_6$ crystal, what is the goal of the present work.

We start from the BEG model supplementing it by the above-mentioned interaction with the lattice deformation (caused both by the external pressure and by the self-consistent changes of occupations of local configurational states). In the framework of this generalization we consider the strain effects appearing in the regions of the first and second order phase transitions to the ferroelectric phase as well as in the vicinity of the tricritical point. In particular, the behaviour of the volume compressibility in the area of the mentioned transitions is analysed.

2. The model

Let us start from the Hamiltonian $\hat{H}$ of the Blume–Emery–Griffiths (BEG) model formulated as a lattice model where three states $|1\rangle$, $|2\rangle$, and $|3\rangle$ with respective energies $E_1$, $E_2$ and $E_3$ are possible for each site. In the representation of Hubbard operators $(X_{i}^{\alpha \beta} = |i, \alpha\rangle \langle \beta, i|)$

$$\hat{H} = \sum_{i} \hat{H}_i + \hat{H}',$$

where the single-site part

$$\hat{H}_i = \sum_{\alpha=1}^{3} E_{\alpha} X_{i}^{\alpha \alpha}.$$

Operators $X_{i}^{\alpha \alpha}$ project on the states $|i, \alpha\rangle$ so their average values $\langle X_{i}^{\alpha \alpha} \rangle$ are equal to occupations of these states. States $|i, \alpha\rangle$ correspond to the

\[ S_i^z = \frac{1}{2}(X_i^{33} - X_i^{22}), \quad n_i = X_i^{33} + X_i^{22}, \]

respectively.

Thus the single-site Hamiltonian can be written down as

$$\hat{H}_i = -\frac{h}{2}(X_i^{33} - X_i^{22}) + E_0(X_i^{33} + X_i^{22}) \equiv -hS_i^z + E_0n_i,$$

where $h$ is the field conjugated to the dipole moment, $E_0 = E_2^{(0)} - E_1 = E_3^{(0)} - E_1$ is the difference of energies of the side and central configurations at $h = 0$ (see figure 1).

In this representation (see [10]) the interaction part $\hat{H}'$ of the Hamiltonian for the BEG model looks like

$$\hat{H}' = -\frac{1}{2} \sum_{ij} J_{ij} S_i^z S_j^z - \frac{1}{2} \sum_{ij} V_{ij} n_i n_j,$$

where $J_{ij}$ and $V_{ij}$ are parameters describing direct dipole and quadrupole interactions between particles. It should be mentioned that for every lattice site a condition $\sum_{\alpha=1}^{3} X_{i}^{\alpha \alpha} = 1$ is fulfilled.

Let us further consider only the dipole interaction $J_{ij}$. We also take into account the possibility of variation of the energy gap $E_0$. In real
locally anharmonic crystals one can achieve this effect applying the pressure: external as well as internal one (caused by stoichiometric substitution of atoms). For example, in the above mentioned crystals of the Sn2P2S6 family the shape of the local three-well potential for ionic groups P2S6 can be changed under the influence of these factors (see ab-initio calculations [5]). Such a behaviour offers insight into the picture of phase transitions to the ferroelectric state.

Taking this effect into account in the framework of the lattice version of the BEG model, one should consider deformation of the crystal lattice (caused by an external factor) as an immediate reason of variation of local potentials (and, thus, the energy gap $E_0$). In this connection the initial Hamiltonian (1) should be supplemented by a respective term

$$\hat{H}'' = D \sum_i (X_{33}^i + X_{22}^i)u + \frac{N}{2} v_0 u^2, \quad (6)$$

taking into account the renormalization of the energy gap $E_0$ due to deformation

$$E_0 \rightarrow \tilde{E}_0 = E_0 + Du \quad (7)$$

(here $u = \Delta V/V$ is a relative change of the volume) as well as the energy of an elastic deformation ($c_0$ is the volume elastic constant, $v$ is the volume related to the one formula unit from the $N$ structure elements described by the locally anharmonic potentials and can be ordered), while $D$ is the constant of an electron-deformational interaction.

A condition of a mechanical equilibrium in the presence of the $\hat{H}''$ interaction can be obtained starting from the condition of a thermodynamic equilibrium $\partial G/\partial u = 0$, where $G$ is the Gibbs free energy derived from the Helmholtz free energy

$$G = F - N v u \sigma, \quad (8)$$

where $\sigma$ is a mechanical stress. Since

$$\frac{\partial G}{\partial u} = \frac{\partial F}{\partial u} - N v u \sigma = \left( \frac{\partial \hat{H}}{\partial u} \right) - N v u \sigma, \quad (9)$$

the condition of a mechanical equilibrium is given by a relation

$$c_0 u + \frac{D}{v} \langle X_{33}^i + X_{22}^i \rangle = \sigma \quad (10)$$

assuming homogeneity of the system (an absence of a modulated ordering).

Condition (10) is an exact relation which can be considered as the generalized Hooke’s law. According to relation (10), the role of the external stress $\sigma$ is not limited only to the deformation of the lattice; the stress also affects the occupations of the side positions ($\bar{n} = \langle X_{22} + X_{33} \rangle$) of local wells. As will be shown below, this results in a non-linear relation between $\sigma$ and $\bar{n}$ what is especially important in the vicinity of the phase transition to the ferroelectric state.

The deformation $u$ can be eliminated using equation (10). As a result, the Hamiltonian of our model can be rewritten as

$$\hat{H} = \sum_i \left[ -h S_i^2 + \left( E_0 + \frac{D}{c_0} \sigma \right) n_i \right] - \frac{1}{2} \sum_{ij} J_{ij} S_i^z S_j^z - \frac{D^2}{v c_0} \sum_i \bar{n} n_i + U, \quad (11)$$

where

$$U = \frac{N}{2c_0} \frac{D^2}{v} - \frac{ND}{c_0} \sigma \bar{n} + \frac{N}{2c_0} \bar{n}^2. \quad (12)$$

The third term in expression (11) has a form corresponding to the energy of a local quadrupole in the mean field $V_{eff} \bar{n}$, where the constant of the effective quadrupole interaction equals to

$$V_{eff} = \frac{D^2}{v c_0}. \quad (13)$$

Such an interaction is a long-range one ($V_{eff} = V_{eff}$ for arbitrary distance $|\bar{R}_{ij}|$) and is mediated by the lattice deformation. However, Hamiltonian (13) is not equivalent to the BEG Hamiltonian with the effective interaction of quadrupoles. The role of the stress $\sigma$ is not limited to the appearance of the field $(D/c_0)\sigma$ acting directly on quadrupoles. It is also manifested in appearance of some additional terms in expression (12). Equivalence to the BEG Hamiltonian is achieved only in the absence of external stresses (at $\sigma = 0$).

### 3. Thermodynamics in the mean field approximation

The model described by the Hamiltonian

$$\hat{H} = \sum_i \hat{H}_i + \hat{H}' + \hat{H}'', \quad (14)$$

can be named as the deformed BEG model (d-BEG). We will consider its thermodynamics in the mean field approximation (MFA). Starting
from definitions (11) and (12), at \( V_{ij} = 0 \) one can obtain
\[
\hat{H}_{MF} = \frac{N}{2} J \eta^2 + \frac{N}{2} v c_0 u^2 + \sum_i \hat{H}_i,
\]
where
\[
\hat{H}_i = (H + \tilde{E}_0) X_i^{22} + (-H + \tilde{E}_0) X_i^{33}.
\]
Here \( H = h/2 + J \eta/2 \) is the effective field acting on dipoles, \( \eta = \langle S_i^z \rangle \) is the parameter of the dipole ordering that determines the polarization of the system. Alongside of deformation \( u \), the “polarization” \( \eta \) belongs to two self-consistency parameters determined by the minimum of the Gibbs free energy \( G \).

Starting from the single-site partition function
\[
Z_i = S_p e^{-\beta \hat{H}_i} = 1 + 2 e^{-\beta \tilde{E}_0} \cosh \beta H,
\]
we obtain the following expression for the Helmholtz free energy
\[
F_{MF} = \frac{N}{2} J \eta^2 + \frac{N}{2} v c_0 u^2 - N \Theta \ln \left(1 + 2 e^{-\beta \tilde{E}_0} \cosh \beta H\right).
\]
Conditions of an extremum of the function \( G_{MF} = F_{MF} - N v u \sigma \)
\[
\frac{1}{N} \frac{\partial G_{MF}}{\partial \eta} = 0, \quad \frac{1}{N} \frac{\partial G_{MF}}{\partial u} = 0
\]
result in this case in the equations
\[
\eta = \frac{e^{-\beta \tilde{E}_0} \sinh \beta H}{1 + 2 e^{-\beta \tilde{E}_0} \cosh \beta H},
\]
\[
c_0 u + \frac{D}{\nu} \frac{2 e^{-\beta \tilde{E}_0} \cosh \beta H}{1 + 2 e^{-\beta \tilde{E}_0} \cosh \beta H} = \sigma.
\]

It is obvious that the second equation coincides with the earlier obtained relation (10) between the deformation \( u \) and the mechanical stress \( \sigma \) thus being the condition of mechanical equilibrium.

Solutions of equation set (20)–(21) for the self-consistency parameters (at given external fields \( h \) and \( \sigma \)) should also correspond to the absolute minimum of the potential \( G \). Such solutions describe thermodynamically stable states (thus omitting metastable and unstable ones) what allows to explore both first and second order phase transitions between various phases.

4. Phase diagrams of the deformed BEG model

Appearance and topology of phase diagrams for the BEG model (regions of existence of the disordered phase, the phase with dipole ordering and phases with various values of the average quadrupole moments) depend substantially on numerical values of model parameters (see, e.g., [11]–[12] as well as [10]). In this connection, we will focus on the particular example of the SnS\(_2\)P\(_2\)S\(_6\) ferroelectric crystal. This crystal was chosen by us for the following reasons: (a) there are attempts to describe thermodynamics of the crystal in the framework of the BEG model [5]; (b) there are data illustrating rearrangement of the local anharmonic potential for ionic groups P\(_2\)S\(_6\) under the influence of an external uniform pressure [8–9].

Starting from the known data for SnS\(_2\)P\(_2\)S\(_6\) [3]–[13], we have fitted the following values of parameters for the deformed BEG model (d-BEG): \( v = 0.23 \cdot 10^{-21} \) cm\(^3\), \( c_0 = 5 \cdot 10^{11} \) erg/cm\(^3\), \( D = -1.1 \) eV, \( \sigma = 71.8 \) eV, \( \tilde{E}_0 = -0.011 \) eV, \( \nu = 0.017 \) eV. The parameter \( D \) was calculated using the definition \( D = \partial \tilde{E}_0/\partial u \) based on the estimation of the derivative \( \partial \tilde{E}_0/\partial u = -\tilde{E}_0/\partial \sigma \approx 0.011 \ldots 0.025 \) eV/GPa according to the results of ab initio calculations [8] (here \( p = -\sigma \) is a hydrostatic pressure). The value of the parameter \( J \) is chosen from the condition of an optimal fit of the critical temperature \( T_c \) at \( p = 0 \), calculated in the framework of the model, comparing to its experimental value \( (T_{c|\exp} = 337 \) K). The value of \( \tilde{E}_0 = -0.011 \) eV corresponds to the data presented in paper [8] for a zero pressure.

The phase diagram \( (T, \tilde{E}_0) \) (figure 2a) at a zero external pressure illustrates a well-known decrease of the critical temperature \( T_c \) at the decrease in absolute value and the subsequent increase in the positive region of the energy parameter \( E_0 \). In the process, the order of the phase transition changes at the tricritical point \( (E_0|\exp \approx 0.02 \) eV) from the second to the first one and the ferroelectric (F) phase is suppressed at \( E_0 \geq 0.026 \) eV.

The dependence of the temperature of the phase transition between the ferroelectric and paraelectric phases \( (F \leftrightarrow P) \) on the applied pressure is presented in figure 2a. Values of the temperatures \( T_c(p = 0) \) and \( T_{TCP} \) as well as the pressure \( p^* \) (at which the temperature of the phase transition tends to zero) are relatively close to the experimental data \( (T_{c|\exp}(p = 0) = 330 \) K, \( T_{TCP|\exp} = 203 \) K and \( p^*|\exp = 1.7 \) GPa while \( T_{c|\exp}(p = 0) = 337 \) K, \( T_{TCP|\exp} = 220 \) K and \( p^*|\exp = 1.5 \) GPa). Hence, such a choice of model parameters can be used as a starting point for a more detailed consideration of the effects induced by an external pressure.
Graphs in figure 3 demonstrate the calculated dependences of the deformation parameter $u$ on the pressure $p$ at various temperatures (below and above of the tricritical point and just at $T = T_{TCP}$). Graphs in figure 4 show the respective dependences of the parameter $\eta$ (describing a spontaneous dipole ordering) on the external pressure (our investigation is limited to the case $h = 0$). The main peculiarity of the curves in figure 3 is a jump of the deformation $\Delta u$ at the first order phase transition from the ferro- (F) to the paraphase (P) accompanied by compression of the lattice. As this takes place, a relative change of the volume $\Delta V/V$ attains to values $-0.011$ corresponding to the measured change of the unit cell volume for the Sn$_2$P$_2$S$_2$ crystal (according to [3], $v_{cell} \approx 0.57 \cdot 10^{-24}$ cm$^3$ for the ferrophase ($T = 293$ K) and $v_{cell} \approx 0.52 \cdot 10^{-24}$ cm$^3$ for the paraphase ($T = 358$ K)).

In the tricritical point $\Delta u \to 0$ and at $T_{c} > T_{TCP}$ the phase transition becomes of the second order, so the jump of $u$ vanishes. However, dependence $u(p)$ still demonstrates a visible peculiarity in the vicinity of the phase transition point. This peculiarity is more pronounced for a respective dependence of the derivative $\partial u/\partial p = -\partial u/\partial \sigma \equiv \chi$ which corresponds to the volume compressibility. The curves, demonstrating dependence of the compressibility $\chi$ on the pressure, are calculated from the equation set (20)–(21) and presented in figure 5. In the tricritical point, the function $\chi(p)$ diverges while nearby this point it demonstrates a peak-like behaviour. Variation of the compressibility within the peak region reaches values of the order of $0.02$–$0.04$ GPa$^{-1}$. An experimental investigation of the compressibility behaviour close to the tricritical

Figure 2. Dependence of the temperature of the ferroelectric-paraelectric phase transition between the ferroelectric (F) and paraelectric (P) phases (a) on the energy parameter $E_0$ (at $p = 0$) and (b) on the applied pressure $p$ (at $E_0 = -0.011$ eV) at the following values of other model parameters: $J = 0.14$ eV, $c_0v = 71.8$ eV, $D = -1.1$ eV, $V_{eff} = 0.017$ eV.

Figure 3. The calculated dependences of the deformation parameter $u$ on the pressure $p$ at various temperatures (174 K, 203 K and 232 K, respectively); here and in figures 4, 5 and 6: $J = 0.14$ eV, $c_0v = 71.8$ eV, $D = -1.1$ eV, $E_0 = -0.011$ eV, $V_{eff} = 0.017$ eV.
Figure 4. The dependences of the “polarization” \( \eta \) on the pressure \( p \) at various temperatures (174 K, 203 K and 232 K, respectively).

Figure 5. The dependences of the compressibility \( \chi \) on the pressure \( p \) at various temperatures (174 K, 203 K and 232 K, respectively).
It should be mentioned that the existence of a mixed phase at $T < T_{TCP}$ was already a subject of discussion [5][9]. Supposedly, an appearance of such a phase is usually related to metastability phenomena in the vicinity of the phase transition between the F and P phases.

5. Conclusions

In the present work we proposed a modified version of the BEG model formulated as a configurational version of a quantum lattice model. The model takes into account the microscopic mechanism of the applied external pressure influence on thermodynamics and phase transitions in crystals with a multi-well (specifically, three-well) local lattice potential. Our approach is based on the idea that the influence of pressure is not a direct one but it is mediated by the crystal lattice strain. Namely, the crystal deformation leads to the change of internal field and displacements of atoms surrounding the structure elements (ionic groups); configurations of the latter are determined by the mentioned above local potential. Our modification of the BEG model supplementarily considers the shift of local energy levels (due to restructuring of local anharmonic potentials formed by the electron subsystem) under the influence of deformation caused by a uniform pressure or tension. Such an approach allows to describe the deformational effects accompanying the phase transitions to the state with a dipole ordering (the ferroelectric phase).

It should be mentioned, that the applied here scheme of implementation of the microscopic parameter (such as deformation) into a macroscopic Hamiltonian with the subsequent determination of such a parameter from the equilibrium conditions was initially proposed in [13]. Based on this approach, the set of effects induced by an external mechanical field in the KH$_2$PO$_4$-type ferroelectrics was described (see [14]).

The Sn$_2$P$_2$S$_6$ crystal is considered as an example. Experimental measurements reveal that the temperature of the ferroelectric phase transition decreases under influence of the hydrostatic pressure with the change of the phase transition order from the second to the first one resulting in a subsequent suppression of the ferroelectric phase at $p = p^*$. This phenomenon is adequately described by the BEG model and the $ab$ $initio$ calculations indicate rearrangement of the local anharmonic potential under pressure. At the same time, we demonstrate in this case that the interaction of energy states of the mentioned structure elements (e.g., groups P$_2$S$_6$ for the Sn$_2$P$_2$S$_6$ crystal) with the lattice deformation, taken into account in the framework of the proposed d-BEG model, leads to the anomaly of the $u(p)$ dependence in the vicinity of the phase tran-

\begin{align}
  x_F &= \frac{u - u_F}{u_p - u_F} \quad x_F = \frac{u_p - u}{u_p - u_F}, \tag{22}
\end{align}

where $x_{P,F}$ are the relative fractions of the P (F) phase, $u_{P,F}$ are the values of the deformation $u$ on the respective boundary of the mentioned interval. Thus, the crystal separates into differently strained fragments of the P and F phases.
sitions from the ferroelectric (F) to paraelectric (P) phase. The lattice compresses and the deformation has a jump $\Delta u$ at the first order phase transition and changes continuously at the second order one. A peak-like behaviour of the $\partial u/\partial p$ function in the vicinity of the phase transition is also revealed. This peak increases approaching the tricritical point where the compressibility $\chi = -\partial u/\partial p$ diverges. Such behaviour of $u(p)$ and $\chi(p)$ coincides with the observed one.

Thermodynamics of $\text{Sn}_2\text{P}_2\text{S}_6$ and the respective phase transitions are also considered in the case of a clamped crystal (regime $u = \text{const}$). As is shown, at $T < T_{TCP}$ the region of deformation values is present in this regime, where the crystal exists in a mixed state being separated into differently strained fragments of the P and F phases. This mixed phase is located between the “pure” P and F phases on the $(T, u)$ phase diagram. Such a state can be experimentally identified by measurement of the compressibility $\chi(u)$ in the “clamped” regime in the area of the mixed state (here $\chi(u)$ is a linear function on $u$ in the interval $[u_F, u_P]$).

References


The journal Condensed Matter Physics is founded in 1993 and published by Institute for Condensed Matter Physics of the National Academy of Sciences of Ukraine.

AIMS AND SCOPE: The journal Condensed Matter Physics contains research and review articles in the field of statistical mechanics and condensed matter theory. The main attention is paid to physics of solid, liquid and amorphous systems, phase equilibria and phase transitions, thermal, structural, electric, magnetic and optical properties of condensed matter. Condensed Matter Physics is published quarterly.

ABSTRACTED/INDEXED IN: Chemical Abstract Service, Current Contents/Physical, Chemical & Earth Sciences; ISI Science Citation Index-Expanded, ISI Alerting Services; INSPEC; “Referaty vnyj Zhurnal”; “Dzherelo”.

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