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Some Static and Dynamic Properties of Non-linear Lattices in Solids

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Abstract: This article describes some results for the static and dynamics of non-linear lattices in solids. First, a non-linear classical lattice with a local fourth order potential is studied for nearest and next nearest neighbour interaction. The effect of interaction coupling is then exemplified for an a-helical model of three chains. An extension to two-dimensional structures is discussed. Dynamical aspects for a non equilibrium situation are presented for the thermal conductivity in a Toda lattice. Finally, a method to describe the effect of electronic correlation on lattice dynamics is presented.

Key words: Lattices, Hamiltonian, Electron- Phonon, Nonlinear.

INTRODUCTION

It is by now well know that one-dimensional lattice models show interesting structural phases non linear excitations and complex dynamical behaviour[7]. In an introductory example we will discuss some of these aspects in a simplified version of a shell model used for the description of ferroelectrics[1] of the phase transition in K_2SeO_4[1] and of the polymorphism in SiO_2,[2]. The special case of three coupled chains as a simple example for a-helical molecules is treated in some detail. These considerations are extended to a real two-dimensional structure with nearest neighbour and next-nearest neighbour interactions Buttner *et al*[2]. The interesting fact is that static structures in two-dimensions are determined by a mapping procedure with two discrete indices. Compared to the one-dimensional models the number of possible structure is greatly enhanced. The application to the surface reconstruction of Silicon is studied. After this discussion of complex static structures in one and two dimensions, a dynamical example is described in detail, namely the diffusion of heat in a diatomic Toda lattice Mokross *et al*[8]. The result of these computer experiments show that the classical Fourier law, namely the linear dependence of the heat current on the temperature gradient, is due to the complex random dynamics of this non-integrable system. Casati *et al*[4].

In all of the above discussion the electrons were treated as classical degrees of freedom in the lattice. In the final analysis we present a method by which a quantum mechanical treatment of the electronic correlation is possible. The method consists mainly of a unitary transformation by which the electronic correlations are eliminated and by which at the same time the electron-phonon interaction is drastically changed.

Theoretical Considerations and Calculations: One dimensional shell models have been used in recent years to study some of the non-linear aspects of lattice dynamics, Bulter *et al*[3].

Especially the description of the phase transition in K_2SeO_4 with a soft phonon near a wave number of two-thirds of the Brillouin zone was a successful application of such a model. In this article we want to introduce some of the complexities in these lattices and instead of discussing in detail the electronic and ionic degrees of freedom we think of only one polarizable constituent in the chain (e.g the SeO_4 complex in K_2SeO_4). Usually, of course, the lattice consists at least of a second unit which may or may not be polarizable (like potassium). However, the dynamics of the second ion within the chain is neglected, it is only used to establish a non-linear potential for the polarizable unit (with polarization V_n). The main feature besides this non-linearity is a competing nearest neighbour (nn) – to next nearest neighbour (nnn) – interaction. The Hamiltonian for the system, treated classically is the Usual V_n- model with competing interactions.

$$H = \sum_n \left[\frac{M}{2} V_n^2 + \frac{f_1}{2}(V_{n+1} - V_n)^2 + \frac{f_2}{2}(V_{n+2} - V_n)^2 + \frac{g_2}{2} V_n^2 + \frac{g_4}{4} V_n^4 \right] \quad (1)$$

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Note that this model is often discussed since the pioneering work of Krumhansl and Schrieffer to describe phase transitions in locally unstable systems ($g_2 < 0$). In actual physical problems one needs a translational invariant model in order to describe also the acoustical modes of excitation for instance by introducing additional electronic degrees of freedom or by using non local nonlinearities. The results we want to discuss actually do not depend on this additional complexity and so we want to describe possible ground states of the above Hamiltonian. Furthermore we concentrate on locally stable lattices by choosing $g_2 > 0$. Especially interesting are the different ground states (for temperature T= 0) as a function of the interaction parameters f_1 f_2. For positive parameters we have the period 1 ground states $V_n = 0$ for all n. This trivial result, however changes if the no interaction become negative and smaller than a certain value. In this case the "paraelectric" state with period 1 changes to a period 2 state with finite displacements at each site a state which might be called anti-ferroelectric. This state is lower in energy since it is favourable to have finite displacements and therefore a contribution from the negative harmonic nn – interaction.

For negative nnn – interactions f_2 the picture changes again. Higher periods become stable because this interaction term gives a negative contribution to the energy. The linear stability analysis shows that for $f_2 < 0$ and $f_1 > 0$ the dispersion of the optical phonon above the period 1 ground state is given by:

$$M\omega_o^2 = g_2 + 2f_1(1 - \cos(ga)) + 4f_2(1 - \cos^2(ga)), \quad (2)$$

and becomes unstable at a certain finite g value determined by

$$\cos(ga) = -f_1/4f_2,\ f_1 = -4f_2 + \sqrt{-4g_2 f_2} \quad (3)$$

The corresponding periodic solution with period $2\Pi/q$ is stable over a finite region of fig1 (with $g_2 = ½$), this is shown for $2\Pi/q = 6a$ with $f_1=1$ and $f_2 = -0.5$). But there are other possible solutions for decreasing f_1 which have much higher periods. The result is quite a complicated phase diagram in this region of parameters. Finally a period 4 patterns results where both the nn – and nnn- interactions are energetically optimized. The parameter line $f_1 = 0$ and $f_2 < 0$ is interesting as it reflects a similar behaviour at the line $f_1 = 0$ and $f_2 < 0$. Only two phases are possible here: the period 1 and the period 4 state for $f_1 = 0$ correspond to the period 1 and period 2 transition for $f_2 = 0$.

As a result one can conclude that even for a harmonically stable non linear potential ($g_2 > 0$) the effect of the competing interactions creates a great number of phases with finite periods. Our numerical study shown in fig 1 has only looked periods up to 23, but of course higher periods are possible and even incommensurable ground states have been found Janssen *et al*[7]. We finally would like to remark that this lattice model is much richer than the Ising model with competing interactions Kaburag, M *et al*[5], since the lattice displacement in our case can have different values for the various periods and therefore has to be determined from the static equation of motion.

Discussing only longitudinal displacements and neglecting transversal components the polarization at site n in chain j is denoted by $Vj,_n$ with j=1, 2, 3, and all n. The corresponding Hamiltonian is then written as

$$H = \sum_{j,n} \left[\frac{M}{2} V_j^2,n + \frac{g_2}{2} V_j^4,n + \frac{f_1}{2}(V_{j,n+1} - V_{j,n})^2 \right]$$
$$+ \frac{f_2}{2} \sum_n \left[(V_{2,n+1} - V_{1,n})^2 + (V_{2,n-1} - V_{1,n})^2 + (V_{3,n-1} - V_{2,n})^2 + (V_{3,n+1} - V_{2,n})^2 + (V_{3,n} - V_{1,n})^2 + (V_{3,n+2} - V_{1,n})^2 \right] \quad (4)$$

where the first chain has been coupled to the second and third, while the second chain is coupled to the first and third chain. Since we expect in a later application the polarizable units to be unstable, we use here a model with $g_2 < 0$. Again of main interest is the instability of the ferroelectric ground state given by:

$$V_j^2,\ n = V_o^2 = -g_2/g_4 \quad \text{for j = 1, 2, 3 and all n} \quad (5)$$

The linearized phonon equations follow from the original equations of motion with the Fourier – ansatz $V_{j,n} = V_o + \Psi j \exp(igan)$. The phonon dispersion are then found from:

$$M^2_\omega \Psi_1 = [-2g_2 + 2f_1(1 - \cos(ga)) + 4f_2]\Psi_1 + f_2[2\cos(ga)\Psi_2 + (1 + e^{-2iga})\Psi_3]$$
$$M^2_\omega \Psi_2 = [-2g_2 + 2f_1(1 - \cos(ga)) + 4f_2]\ \Psi_2 + 2f_2[2\cos(ga)\Psi_3 + \cos(ga)\ \Psi_1]$$
$$M^2_\omega \Psi_3 = [-2g_2 + 2f_1(1 - \cos(ga)) + 4f_2]\ \Psi_3 + f_2[2\cos(ga)\Psi_2 + (1 + e^{2iga})\Psi_1] \quad (6)$$

These equations can be solved exactly for the three optical modes. The result is:

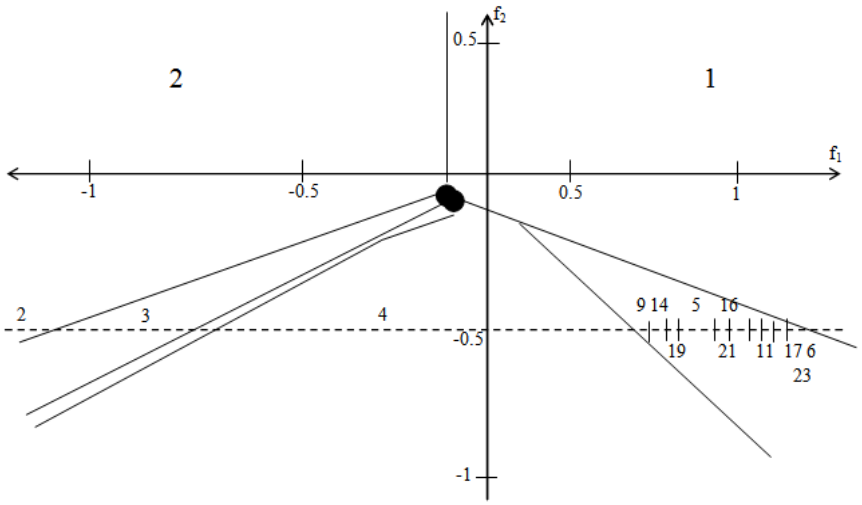

Fig. 1: Commensurate ground states of the one dimensional model as function of the parameters f_1 and f_2 for g_2= ½ and g_4= ½. Along the broken line the states with lowest energy are shown up to period 23. In the shaded area no numerical results are available.

$$M^2\omega_1 = -2g_2 + 2f_1(1 - \cos(ga)) + 4f_2(1 - \cos(ga)\cos(ga/3/)$$
$$M^2\omega_{2,3} = -2g_2 + 2f_1(1 - \cos(ga)) + 2f_2[2 - \cos(ga)\cos(ga/3/) \pm \sqrt{3}\sin \quad (ga/3)] \qquad (7)$$

A quantitative example is shown in fig 2 for $f_1 > 0$ and two different $f_2 < 0$ in the extended Brillouin zone. The minimum of the dispersion curve is always near ga= Π/j_o and quite insensitive to the detailed ratio f_1/f_2. The only necessary condition is the negative sign of f_2.

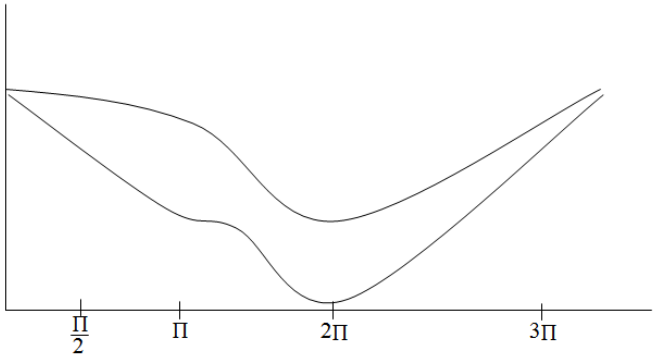

Fig. 2: Optical phonon dispersion (in the extended zone scheme) above the ground state $V_o^2 = -g_2/g_4$ for fixed g_2, g_4 and f_1 and two different $f_2 < 0$. The minimum near 2p indicates the instability.

The reciprocal value of the instability point gives the period of the new structure p= 20a, this value is remarkably close to the period found in most a- helical structure Davydov and Scolt (which is 5 x 3.6 where one has 3.6 units per term). This possible application of the above model to the a- helix is quite realistic since in this structure one has highly polarizable units (the c= 0 - - -H- N- complex) and also three coupled, twisted chains. In a first approximation one may neglect the other carbon ions and treat them as an effective coupling mechanism. They will of course, influence the strength of the interaction and also their signs, but it is quite interesting that our calculation shows that the resulting periodic structure is to extend insensitive to the details of the interaction.

We are well aware of the fact that the treatment of the a- helix by Davydov and Scott uses dipolar interactions to describe non linearities and also a quantum – mechanical description of the hydrogen band. These authors were mainly interested in describing the excitations in these structures, while our lattice phenomenological example shows how the ground- state in the structure may be explained by polarization forces that are quite similar to the original proposal. We expect the lattice excitations to have also a very similar behaviour.

The above discussion is extended to the case of a two- dimensional hexagonal structure. The main point we want to stress here is the fact that the mappings corresponding to the static pattern have two discrete indices and differ therefore from all known mappings of dynamical systems (with one time index) or one-dimensional lattices (with one space point index). Our example treats some of these aspects in a simple hexagonal lattice with nn- and nnn- interactions. It is the simplest model which can also serve for the description of surfaces and other layered structures. The variety of the periodic solutions is greatly enhanced compared to the one- dimensional chain.

The corresponding Hamiltonian is given in the following form (where non orthogonal lattice vectors are used).

$$H = \sum_{n,m} \left[\frac{M}{2} V_{n,m}^2 + \frac{f_1}{2}[(V_{n.m} + V_{n,m+1})^2 + (V_{n,m} - V_{n+1,m})^2 + (V_{n,m} - V_{n+1,m-1})^2] \right.$$
$$\left. + \frac{g_2}{2} V_{n,m}^2 + \frac{g_4}{4} V_{n,m}^4 + \frac{f_2}{2}[(V_{n,m} - V_{n+1,m+1})^2 + (V_{n,m} - V_{n+2,m+1})^2 + (V_{n,m} - V_{n+1,m-2})^2] \right] \quad (8)$$

with the parameters g_2 and g_4 describing the non linear onsite potential. From the corresponding classical equations of motion one easily finds the mapping relations for the static structures.
These relations are given by:

$$0 = f_1\{ V_{n,\,m-1} + V_{n,\,m+1} + V_{n+1,\,m-1} + V_{n+1,\,m} + V_{n-1,\,m+1} + V_{n-1,\,m} - 6V_{n,\,m}\} + f_2\{V_{n+1,\,m-2} + V_{n+1,\,m+1} + V_{n-1,\,m-1} + V_{n-1,\,m+2} + V_{n+2,\,m-1} + V_{n-2,\,m+1} - 6V_{n,\,m}\} - g_2 V_{n,\,m} - g_4 V_{n,\,m}^3 \quad (9)$$

Formally the mapping allows to calculate e.g. the value of $V_{n-1,\,m+2}$ if all other values in the mapping relation are given. This case is represented in fig 3, where all values on the starting lines m= 1, 2, 3, 4 have to be known in order to determine the lattice displacements on line 5. From this, one can then proceed to determine the values for line 6, and so on. Note, however, that the boundary values needed for the construction of the two- dimensional lattice constitute a one- dimensional set.

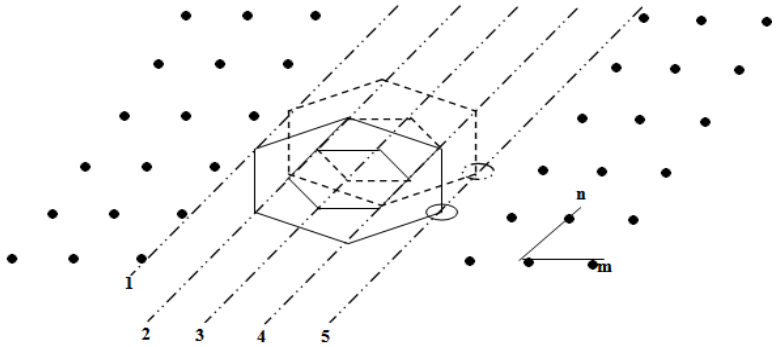

Fig. 3: Geometry for the mapping in two dimensions. The encircled points are the results of the recursion-relation for two different starting configurations: full and broken lines on lines 1, 2, 3, and 4.

The most interesting areas in the phase diagram is again the instability curve of the period/ structure. It is found as in the above examples from the phonon dispersion (for $g_2 > 0$, a locally stable lattice).

$$M_\omega^2 = g_2 + 4f_1\{4 - \cos(\rho a/2)[\cos(\sqrt{3}ga/2) + \cos(\rho a/2)]\} + 4f_2\{4 - \cos(\sqrt{3}ga/2) + \cos(\sqrt{3}ga/2 + \cos(3\rho a/2))]\}, \quad (10)$$

where g, p_o are the wave numbers in the reciprocal lattice. The instability occurs whenever $M\omega^2 = 0$ at a certain (g, p) point. For a given set of parameters the exact transition has to be determined numerically.

The above model is also related to the so called layer model where periodic variations are only allowed in one direction of the lattice. In this case one assumes that the static displacements are independent of the index n, that means $V_{n,\,m} = V_{n+1,\,m} = V_m$. The static mapping in this case is then equivalent to a one-dimensional lattice with some what redefined coupling constants.

$0= (2f_1 + 2f_2)(V_{m+1} + V_{m-1} - 2V_m) + f_2(V_{m+2} + V_{m-2} - 2V_m) - g_2V_m - g_4V_m^3$ (11)

RESULTS AND DISCUSSIONS

In the above discussions on one-and two-dimensional lattices the electronic degrees of freedom were only taken into account by an effective onsite potential. Even in the treatments of a shell model the electrons are treated as classical particles neglecting their fermion character. For any applications, however, its important to study quantum mechanical aspects of the electronic contributions. A phenomenological model which includes the electron – lattice coupling as well as the correlations is a peierts system with additional Hubbard – like correlations. In the following we will treat this model by describing a method by which the correlation term may be eliminated at the expense of an effective electron-phonon coupling Weber and Buttner. The model Hamiltonian is written as:

$$H = \sum_n \left[\varepsilon_1 n_1 + (\frac{\rho_t^2}{2M} + \frac{f}{2}u_1^2) + \alpha n_1 u_1 + Jn\uparrow n_1\downarrow \right] - t_o \sum_{1,s} (C_{e+1,s}C_{1,s} + h.c) \quad (12)$$

which is quite similar to that used by Holstein for molecular crystals (besides the correlation term). While l_o describe the electronic hopping, the parameter a is the electron-lattice coupling constant and j denotes the electronic correlation strength. The electronic density at site l, $n_l = n_l\uparrow + n_l\downarrow$, is the sum of the different spin contributions. We do not intend to establish an analogy to the classical shell model treatment but we want to show how the correlations can be treated effectively. The main idea is an unitary transformation which displaces the lattice oscillators by an amount which is dependent on the electronic density. The transformation operator maybe written in the following form:

$$T = \pi_1 \exp\{\frac{i}{h}\sum_S (A_{1.s}U_1 - B_{1,s}P_1)n_{1,s}\} \quad (13)$$

with variational parameters $A_{l,\ s}$ and $B_{l,\ s}$. the corresponding transformations of the different operators are given by:

$$TU_1T^+ = U_1 - \sum_S B_{1,s}n_{1,s}$$
$$TP_1T^+ = P_1 - \sum_S A_{1,s}n_{1,s} \quad (14)$$
$$TC_{1,s}^+T^+ = \exp\{\frac{\imath}{h}(A_{t,s}U_1 - B_{1,s}P_c)\}C_{1,s}^+$$

Note that the displacements in U_l and P_l are now dependent on the electronic densities and therefore describe Polaron-like quantities. The resulting transformed Hamiltonian follows directly:

$$H = H_o - t_o\sum_{t,s}\left[\exp\{-\frac{\imath}{h}(B_{l+1,s}P_{l+1} - B_{1,s}P_1)\} - 1\right]C_{l+1,s}^+C_{1,s} + hc + \sum_{t,s}\left[\frac{f}{2}(B_{1,s} - 2U\imath) - \alpha\right]B_{1,s}n_{1,s} \quad (15)$$

The two additional terms in this expression describe a phase change in the hopping term and a refined electron-phonon interaction. In a special choice of the variational parameters we use $A_{l,\ s} = 0$, and in order to get rid of the correlations the parameters B have to fulfil the following condition at each site:

$fB_l\uparrow \ . \ B_l\downarrow \ - \ a(B_l\downarrow \ . \ B_l\uparrow) + J = 0$ (16)

Conclusion: Together with the appropriate ansatz for the ground state one has to find an optimal set of parameters B. Examples discussed above show that a possible wave function for the ground state can be written as a product of a Boson-contribution |B> and a fermion part |F>. For |B> a coherent – (or Glauber-) state was used:

$$|B\rangle = \Pi_l \exp\left(-\frac{\iota}{h}\ (z_l P_l - X_l U_l)\right)|0\rangle_B \qquad (17)$$

with additional parameters Z_l, X_l. B_l, this ansatz the Boron-operators maybe treated as classical variables. The method was successfully tested for the two-particle two site problem as well as system with up to 24 particles.

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