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Nonlinear dynamics of a two-dimensional lattice

NABONITA D CHOWDHURY and SANKAR P SANYAL
Department of Physics, C V Raman Building, Barkatullah University, Bhopal 462026, India

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Abstract. The dynamics of the nonlinear excitations in a two-dimensional (2D) ϕ^4-diatomic lattice, with nonlinear on-site electron–phonon coupling at the polarizable ion site has been presented, without considering the self consistent phonon approximation. One of the major results obtained from our calculations is in the understanding of continuous structural phase transition, where we have obtained the minimum in soft mode frequency at a soft mode temperature $T_s(> T_c)$, not at critical temperature T_c. This occurs due to the anisotropy of such 2D systems.

Keywords. Lattice dynamics; phase transition; soft mode; nonlinear dynamics.

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1. Introduction

During the last decade a great deal of interest has emerged to understand the dynamics of one-dimensional diatomic lattices with nonlinear ϕ^4-polarization potential at the anion site [1, 2]. The ϕ^4-polarizability model successfully describes the displacive type of phase transition in solids and its dynamics strongly resembles the structural phase transition in ferroelectrics and ferromagnetic materials. The phenomenology behind the structural phase transition (SPT) is that, the system goes from a stable high temperature phase to low temperature phase, at some critical temperature T_c at which, the frequency of a vibrational mode reduces to zero, a phenomenon well known as "mode softening" [3]. The motivation for studying the weakly coupled two-dimensional (2D) chains in the present paper is stronger than of academic interest.

Also the quasi one-dimensional materials e.g., polyacetylene toluene sulfonate [4] and low dimensional magnetic materials [5], which undergo mode softening and structural inhomogeneities near the transition have drawn considerable interest. Looking at the experimental successes in the identification and isolation of strongly nonlinear effects [4, 5], it is time to develop more theoretical understanding of the two-dimensional materials. Horovitz *et al* [6] for the first time investigated the solitary solutions in a two-dimensional lattice with inter-chain coupling. Static solitary excitations for the 2D lattice having strong anharmonic polarizability at the anion have been obtained by Behnke and Buttner [7]. This type of lattice serves as a model for certain ferroelectric materials. They found that the solitary finite energy excitations are stable against small perturbations. Apart from static solitary excitations, nonlinear periodic solutions are also obtained with harmonic first and second nearest neighbour interactions [7]. Kerr and Bishop [8] have investigated the central peak phenomena at

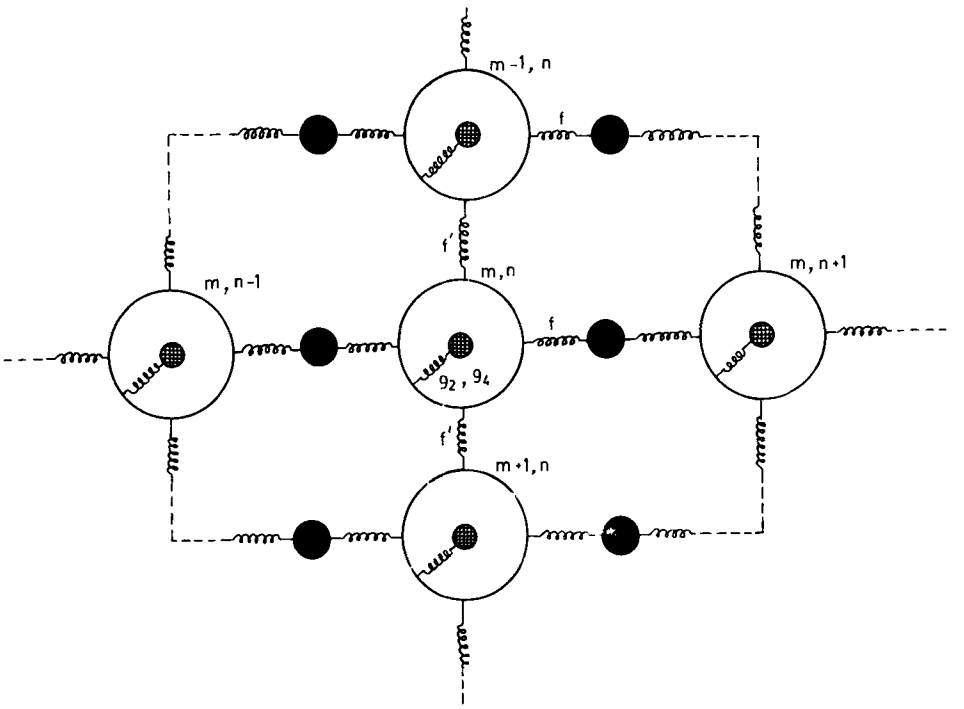

Figure 1. A two-dimensional lattice with on-site nonlinear electron-phonon coupling at the polarizable ion.

SPT using computer simulation technique for a two-dimensional anharmonic lattice. They have used a model of displacive ϕ^4-monoatomic chain which is coupled weakly in a 2D array with nonlinear interparticle coupling and predicted the incomplete mode softening at the transition temperature.

In the present paper we aim to investigate the dynamics of the nonlinear excitations in two-dimensional displacive diatomic lattice with an on-site fourth order anharmonic electron–phonon coupling at the polarizable ion site. Our analytical treatment differs from the previous workers [6–8], in the sense that we consider the nonlinearity in the core-shell coordinate of the polarizable ion (i.e. on-site). We have also not considered the mean field approximation for obtaining the results as reported earlier [6, 7]. We have calculated the frequency and energy of the 2D anharmonic oscillators and discussed the results in the light of the motion of domain walls in ferroelectric materials.

2. Theory

The main features of the present 2D lattice with on-site nonlinear electron-phonon interactions of fourth order at the polarizable ion site can be seen from figure 1. It consists of m linear diatomic chains along x-axis of the lattice with a highly localized fourth order core-shell coupling at the anion site and a rigid ion at the cation site. These diatomic chains are coupled by a linear nearest neighbour shell-core force constant (f)

along the x-axis and nearest neighbour shell–shell force constant (f') along the y-axis. Here the different chains are depicted by m and the different ions in the chain are counted by n. Also we assume that the strength of the coupling along the x-axis is larger in magnitude, as compared to the coupling along the y-axis. Thus it reduces to an effective one-component displacement model in a 2D diatomic lattice, resulting into weak anisotropy.

The displacement of the core of the anion at the nth position along the mth chain from its lattice site is denoted by $u_1^{m,n}$. Other displacements of the cation and shell of the anion are represented as $u_2^{m,n}$ and $v^{m,n}$ respectively (see figure 1). The polarization potential of the anions in 2D displacive array can be written as

$$\phi_P = \frac{1}{2}\sum_{m,n}\left[g_2(v^{m,n}-u_1^{m,n})^2 + \frac{1}{2}g_4(v^{m,n}-u_1^{m,n})^4\right] \tag{1}$$

where g_2 and g_4 are the on-site shell-core quartic (harmonic) and biquadratic (anharmonic) force constants. Here $g_2(<0)$ and g_4, f (both >0) are the parameters describing the double-well potential at (m,n)th site. This ϕ^4-potential has a minima at the displacements $w^{m,n} = \pm(-g_2/g_4)^{1/2}$ [1, 2]. The polarization potential for this displacement is obtained as $(-g_2^2/4g_4)$.

The Hamiltonian for the two-dimensional diatomic lattice can be written as

$$\begin{aligned} H = {} & \frac{1}{2}\sum_{m,n}[m_i(\dot{u}_i^{m,n})^2 + m_{ej}(\dot{v}_j^{m,n})^2] \\ & + \frac{1}{2}\sum_{m,n}[f(v^{m,n}-u_2^{m,n})^2 + f(v^{m,n}-u_2^{m,n-1})^2 \\ & + f'(v^{m+1,n}-v^{m,n})^2 + f'(v^{m-1,n}-v^{m,n})^2] + \phi_P \\ & i = 1,2 \quad j = 1. \end{aligned} \tag{2}$$

In order to obtain physical solutions for the 2D anisotropic lattice, the excitations are assumed to be propagating slowly in the x and y directions. Hence, it is appropriate to use the continuum limit, in which the relative shell-core displacement $w^{m,n}$ $(= v^{m,n} - u_1^{m,n})$ can be written as [7]

$$\begin{aligned} w^{m,n} &= w^m(x) = w(y,x) \\ w^{m\pm1,n} &= w^{m\pm1}(x) = w(y\pm a_2, x) \\ w^{m,n\pm1} &= w^{m\pm1}(x\pm a_1) = w(y, x\pm a_1) \end{aligned} \tag{3}$$

Similar solutions are also assumed for the displacements $u_1^{m,n}$ and $u_2^{m,n}$. Here a_1 and a_2 are the lattice spacings in x and y directions respectively. The relative displacements of the neighbouring two polarizable ions can be expanded for small lattice spacing a_1 and a_2 in Taylor's series as [7]

$$w(y, x\pm a_1) = w(y,x) \pm a_1 w_x(y,x) + \frac{a_1^2}{2}w_{xx}(y,x) \tag{4}$$

$$w(y\pm a_2, x) = w(y,x) \pm a_2 w_y(y,x) + \frac{a_2^2}{2}w_{yy}(y,x) \tag{5}$$

where $w_x(y,x) = (\mathrm{d}w(y,x))/\mathrm{d}x$ and $w_{xx}(y,x) = (\mathrm{d}^2 w(y,x))/\mathrm{d}x^2$ are the first and second derivatives with respect to x. Similar expressions can also be obtained for y direction.

Using the adiabatic approximation ($m_{ej} \sim 0$) and the method described in [2], we have obtained a coupled nonlinear equation of motion in terms of relative core-shell coordinate of the anion as

$$\begin{aligned}
&[2f + g_2 + 3g_4(w^{m,n})^2]\ddot{w}^{m,n} + [g_2 + 3g_4(w^{m+1,n})^2]\ddot{w}^{m+1,n} \\
&\quad + [g_2 + 3g_4(w^{m-1,n})^2]\ddot{w}^{m-1,n} + 6g_4[(\dot{w}^{m+1,n})^2 w^{m+1,n} \\
&\quad + (\dot{w}^{m-1,n})^2 w^{m-1,n} + (\dot{w}^{m,n})^2 w^{m,n}] + \frac{f}{\mu}[g_2 w^{m,n} + g_4(w^{m,n})^3] \\
&\quad + \frac{f}{m_2}[g_2 w^{m+1,n} + g_4(w^{m+1,n})^3] + \frac{f}{m_2}[g_2 w^{m-1,n} + g_4(w^{m-1,n})^3] = 0 \qquad (6)
\end{aligned}$$

with

$$\frac{1}{\mu} = \frac{1}{m_1} + \frac{1}{m_2}$$

and

$$\begin{aligned}
f'(v^{m+1,n} + v^{m-1,n} - 2v^{m,n}) &= g_2(w^{m+1,n} + w^{m-1,n}) \\
&\quad + g_4[(w^{m+1,n})^3 + (w^{m-1,n})^3]. \qquad (7)
\end{aligned}$$

In this paper we have considered the solutions for those nonlinear excitations which are uniformly varying along y direction i.e., $w_y (= \mathrm{d}w/\mathrm{d}y)$ is constant, and hence the second derivative w_{yy} $(= \mathrm{d}^2 w/\mathrm{d}y^2)$ reduces to zero. In view of this fact, the relative core-shell displacement depends linearly along y direction (i.e., $w = cy$). By using (3), (4), (5) and the above assumptions, the second order differential equation of motion (eq (6)) reduces to

$$\begin{aligned}
&(\alpha + \beta' w^2 + \frac{2}{3}\beta' a_2^2 w^2)\ddot{w} + 2\beta' \dot{w}^2 w + \frac{f}{\mu}(g_2 w + g_4 w^3) \\
&\quad + \frac{2}{9}\beta' g_4 a_2^3 w_y^3 + \frac{2}{3}\beta' \frac{g_2}{m_2} a_2^2 w_y^2 w = 0 \qquad (8)
\end{aligned}$$

where

$$\alpha = 2f + g_2 \quad \text{and} \quad \beta' = 9g_4.$$

The method of solving such types of differential equation has been given in detail in [2]. The first integration of (8) using boundary conditions results into

$$\dot{w}^2 = \frac{f}{27\mu} \frac{F(w)}{\left(w^2 + \frac{\alpha}{\beta'} + \frac{2}{3}a_2^2 w_y^2\right)^2} \qquad (9)$$

where

$$\begin{aligned}
F(w) &= w^6 + w^4\left[\frac{3}{2}\left(\frac{\alpha}{\beta'} + \frac{g_2}{g_4}\right) + a_2^2 w_y^2\left(1 + \frac{9\mu}{m_2}\right)\right] + \frac{6\mu}{m_2} a_2^3 w_y^3 w^3 \\
&\quad + \left(3\frac{g_2}{g_4} + \frac{9\mu}{2m_2} a_2^2 w_y\right)\left(\frac{\alpha}{\beta'} + \frac{2}{3}a_2^2 w_y^2\right) w^2
\end{aligned}$$

$$+\frac{18\mu}{m_2}a_2^3 w_y^3\left(\frac{\alpha}{\beta'}+\frac{2}{3}a_2^2 w_y^2\right)-\lambda \tag{10}$$

C_0 and λ are the constants of integration and are defined in terms of initial root of the displacement w_0. The frequency is obtained from (9) by integrating it numerically and made temperature dependent by using the formula

$$T=\frac{\mu}{NK_B}\sum_n \dot{w}_{1,n}^2. \tag{11}$$

The energy of the system at the turning point ($w^{m,n}=\pm w_0$) can be obtained as

$$E=\frac{1}{2f}[(u_2^{m,n}-v^{m,n})^2+(u_2^{m,n-1}-v^{m,n})^2]$$

$$+\frac{1}{2f'}[(v^{m+1,n}-v^{m,n})^2+(v^{m-1,n}-v^{m,n})^2]$$

$$+\frac{1}{2}g_2(w^{m,n})^2+\frac{1}{4}g_4(w^{m,n})^4. \tag{12}$$

The energy expression is simplified by using (3), (4), (5) and (7). The energy is obtained as a sixth order polynomial in w_0, and gives an asymmetric double-well with respect to displacement with a central hump shifted towards the less equilibrium state, as discussed in the next section.

3. Results and discussion

The values of the parameters are chosen as $g_2/g_4=-0{\cdot}2$, $\alpha/\beta'=0{\cdot}2$, $\mu/m_2=0{\cdot}1$ and $a_2w_y=0{\cdot}02$ in order to get a double-well structure of the potential, so that the scope of the present analysis includes simple ferroelectric systems. The force constants f, $g_2(<0)$ and f' are in the unit of 10^4 g/s^2 and $g_4(>0)$ is in the unit of 10^{22} g/(cm^2 s^2). The above values of the parameters are chosen, keeping in mind the conditions $|g_2|\ll g_4$ and $f<|g_2|$ to be obeyed for a double-well potential. The parameter μ/m_2 is a dimensionless quantity. The reduced mass μ is taken to be small as compared to the unpolarizable ion at (m,n)th site since the unpolarizable ion has been assumed to be heavier and has less displacement as compared to the polarizable ion. The parameter a_2w_y is in unit of lattice constant. The small value of the parameter a_2w_y is sufficient enough to visualize the dynamics of the anisotropic two-dimensional array.

The energy of the two-dimensional (2D) lattice as a function of initial displacement w_0 is shown in figure 2. An asymmetric double-well structure of the energy is obtained with a central hump shifted to one side and crosses the displacement axis at two different points. This fact decides the appearance of two characteristic temperatures, the critical temperature T_c and soft mode temperature T_s in an anisotropic two-dimensional lattice, similar to the one already shown by Bishop [8] who has carried out molecular dynamics of weakly coupled 2D lattice.

In figure 3 we have plotted the square of the soft mode frequency as a function of dimensionless temperature ($NK_BT/(g_2^2/g_4)$). From this figure, it is evident that the minimum in the soft mode frequency does not occur at the transition temperature T_c,

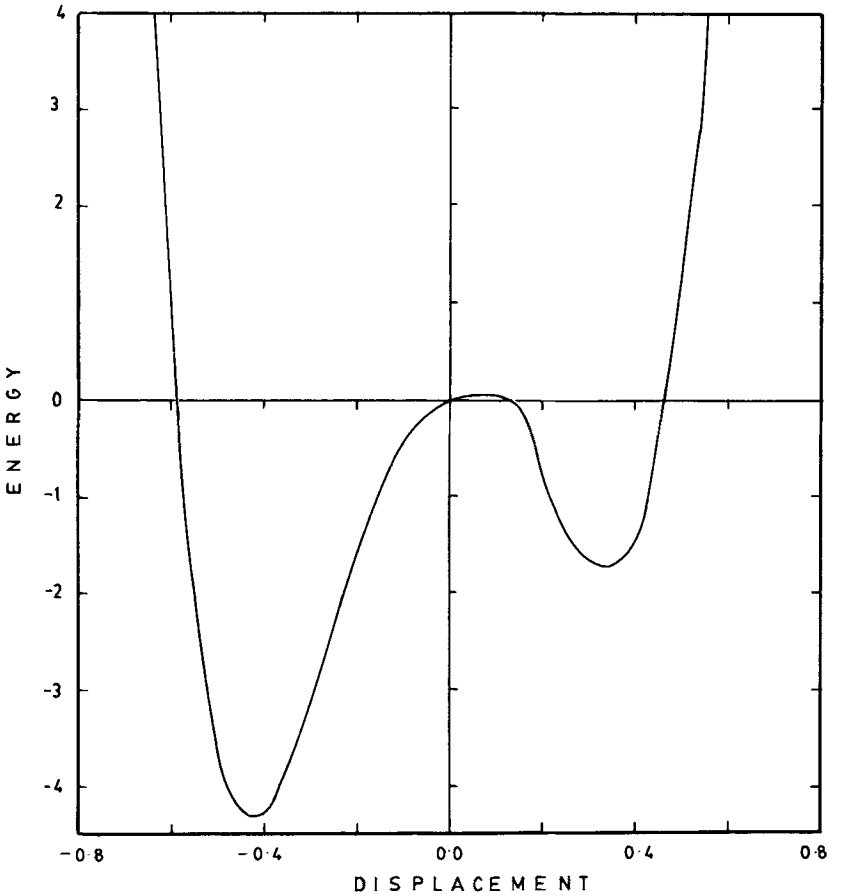

Figure 2. Energy of the two-dimensional lattice is plotted as a function of initial displacement w_0.

but at a temperature slightly greater than T_c, called soft mode temperature T_s. For temperatures much above T_s, the oscillations are completely across the asymmetric double-well resulting into an increase in frequency. In the temperature regime $T < T_s$, the particle can oscillate in either of the wells depending upon the equilibrium energy state (see figure 2). At very low temperature, the particle oscillates in either of the wells and remain localized leading to harmonic oscillation in this temperature regime ($0 < T < T_c$). Instability in the frequency at T_c is obtained because of the reduction of the domain wall separation between the para- and ferro-electric phases, which increase the electron-phonon interactions in the 2D system. Complete minimum does not occur at T_c as there remains some localized region in one of the equilibrium states. As a result, the soft mode frequency increases for a while with displacement and then reduces to its minimum at T_s. The reduction of frequency at T_s is very sharp which indicates that one will obtain a singularity in the physical properties at T_s (and not at T_c) for such a weakly coupled two-dimensional systems.

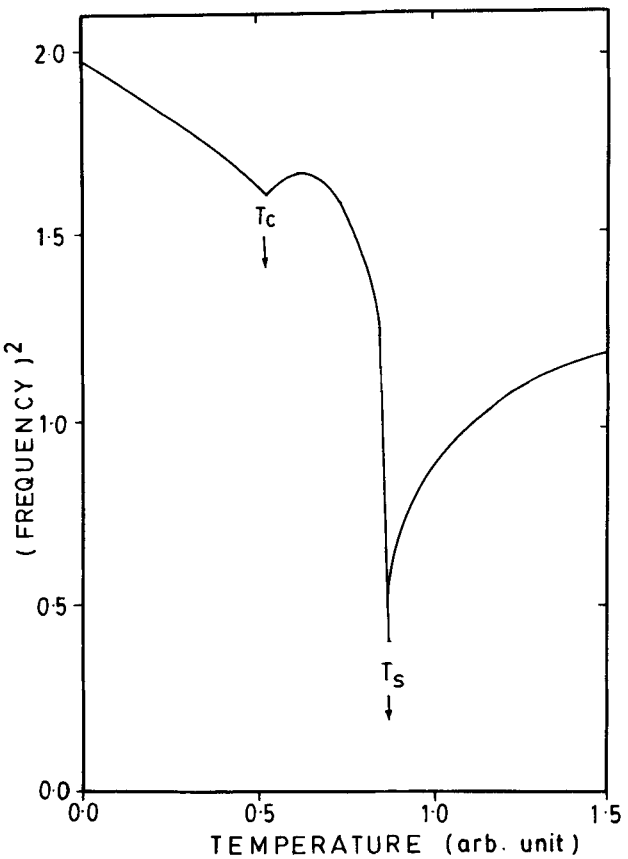

Figure 3. Square of the frequency of the 2D lattice versus dimensionless temperature ($NK_BT/(g_2^2/g_4)$).

Similar modelling of two-dimensional lattice with different types of ions at the alternate positions in the same column can be made, which leads to a somewhat simplified picture as the force constants between the shell of polarizable ion and core of cation along the chain and perpendicular to the chain will become nearly the same. This type of modelling of 2D lattice leads to only one transition temperature T_c, as the equation of motion is isotropic with respect to core-shell displacement of polarizable ion and hence no mode softening temperature T_s.

Our results have indicated that anisotropic 2D lattice (figure 1) shows interesting behaviour at the structural phase transition. These types of studies will help in the understanding of many recent experiments on the structural phase transition of hydrogen bonded macro-molecules [9] and quasi one-dimensional materials [10].

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