Phonons in broken-symmetry structures

Bratislav S. Tošić and Jovan P. Šetrajčić
Institute of Physics, Faculty of Sciences, University of Novi Sad, 21000 Novi Sad, Yugoslavia

Rajka P. Djajić-Jovanović
Institute of Applied Fundamental Disciplines, Faculty of Technical Sciences, University of Novi Sad, 21000 Novi Sad, Yugoslavia

Dragoljub Lj. Mirjanić
Faculty of Technology, University "Djuro Pucar Stari" of Banja Luka, 78000 Banja Luka, D. Mitrova 63b, Yugoslavia
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A parabolic model of structure deformation is proposed. This model can be realized with a specific pressure distribution over the boundaries of a film. The parabolic deformation, if achieved, causes the appearance of optical phonons only. It is shown that the population of optical phonons rapidly decreases with decreasing film thickness.

I. INTRODUCTION

There has been increasing interest in phonon physics over the past few years. Different types of broken symmetry as well as their influence on relevant physical characteristics have been the object of intensive theoretical[1-5] as well as experimental investigations.[6-10] In order to contribute to these above-mentioned efforts we shall propose a model of structure deformation and discuss the expected effects on phonon characteristics.

We shall analyze phonons in the simple-cubic structure with broken symmetry along the z axis. The translation symmetry is conserved in planes orthogonal to the z axis. In order to simplify calculations we assume that the torsion coefficients $C^{\alpha\beta}$, $\alpha\neq\beta$, $\alpha,\beta=(x,y,z)$ are equal to zero. Under the above assumptions the vibrational Hamiltonian of the system can be written in the form

$$H=\sum_{\mathbf{n},\beta}\frac{1}{2M_{\mathbf{n}}}(p_{\mathbf{n}}^{\beta})^2+\tfrac{1}{4}\sum_{\mathbf{n},\boldsymbol{\lambda},\beta}C_{\mathbf{n}}^{\beta\beta}(\boldsymbol{\lambda})(U_{\mathbf{n}}^{\beta}-U_{\mathbf{n}-\boldsymbol{\lambda}}^{\beta})^2 . \tag{1}$$

In this formula $M_{\mathbf{n}}$ are the masses of atoms, $U_{\mathbf{n}}^{\beta}$ are their displacements, $p_{\mathbf{n}}^{\beta}=M_{\mathbf{n}}\dot{U}_{\mathbf{n}}^{\beta}$ are momenta, $\boldsymbol{\lambda}$ is the vector connecting the nearest neighbors in a simple-cubic lattice, and $C_{\mathbf{n}}^{\beta\beta}(\boldsymbol{\lambda})$ are the strain coefficients. The breaking of symmetry is taken into account by taking M and C to be dependent upon the lattice point vector $\mathbf{n}$. The explicit form of the functions $M_{\mathbf{n}}$ and $C_{\mathbf{n}}$ will be chosen later.

II. DISPLACEMENT EQUATIONS

Using the equations of motion

$$i\hbar\dot{p}_{\mathbf{f}}^{\alpha}=[p_{\mathbf{f}}^{\alpha},H],\quad i\hbar\dot{U}_{\mathbf{f}}^{\alpha}=[U_{\mathbf{f}}^{\alpha},H] , \tag{2}$$

and taking $p_{\mathbf{f}}^{\alpha}(t)=p_{\mathbf{f}}^{\alpha}(0)e^{-i\omega t}$ and $U_{\mathbf{f}}^{\alpha}=U_{\mathbf{f}}^{\alpha}(0)e^{-i\omega t}$, where ω is the frequency, it is easy to obtain the following equation defining atom displacements in a structure with the Hamiltonian (1):

$$\omega^2U_{\mathbf{f}}^{\alpha}=\frac{1}{2M_{\mathbf{f}}}\sum_{\boldsymbol{\lambda}}C_{\mathbf{f}}^{\alpha\alpha}(\boldsymbol{\lambda})(2U_{\mathbf{f}}^{\alpha}-U_{\mathbf{f}+\boldsymbol{\lambda}}^{\alpha}-U_{\mathbf{f}-\boldsymbol{\lambda}}^{\alpha}) . \tag{3}$$

Taking into account that the translation symmetry is broken only along the z axis, one can write

$$\mathbf{f}\equiv(f_x,f_y,f),\quad -\frac{N_x}{2}\le f_x\le\frac{N_x}{2},\quad -\frac{N_y}{2}\le f_y\le\frac{N_y}{2},\quad 0\le f\le N$$
$$M_{f_x,f_y,f}\equiv M_f,\quad C^{\alpha\alpha}_{f_x,f_y,f;f_x,f_y,f\pm1}\equiv C^{\alpha\alpha}_{f,f\pm1} , \tag{4}$$
$$C^{\alpha\alpha}_{f_x,f_y,f;f_x\pm1,f_y,f}=C^{\alpha\alpha}_{f_x,f_y,f;f_x,f_y\pm1,f}\equiv D_f^{\alpha\alpha} ,$$

where N_α is the number of atoms along the α direction. After substitution

$$U^{\alpha}_{f_x,f_y,f}=U_f^{\alpha}\cos(f_xk_x+f_yk_y)a,\quad \mathbf{k}\equiv(k_x,k_y) ,$$
$$k_x=\frac{2\pi}{N_xa}\nu_x,\quad k_y=\frac{2\pi}{N_ya}\nu_y,\quad \nu_x\in\left[-\frac{N_x}{2},\frac{N_x}{2}\right],\quad \nu_y\in\left[-\frac{N_y}{2},\frac{N_y}{2}\right] , \tag{5}$$

where a is the lattice constant and accounting for (4), Eq. (3) reduces to the following set of equations:

$$U_{f+1}^{\alpha}+U_{f-1}^{\alpha}Q_f^{\alpha\alpha}U_f^{\alpha}=0, \quad f=1,2,3,\ldots,N-1 \tag{6}$$

$$U_1^{\alpha}-Q_0^{\alpha\alpha}U_0^{\alpha}=0, \quad f=0 \tag{7}$$

$$U_{N-1}^{\alpha}-Q_N^{\alpha\alpha}U_N^{\alpha}=0, \quad f=N\ . \tag{8}$$

The functions Q figuring in (6)–(8) are given by

$$Q_f^{\alpha\alpha}=2\left[1-\frac{M_f\omega^2-4D_f^{\alpha\alpha}[\sin^2(k_xa/2)+\sin^2(k_ya/2)]}{C_{f,f+1}^{\alpha\alpha}+C_{f,f-1}^{\alpha\alpha}}\right], \tag{9}$$

$$Q_0^{\alpha\alpha}=2\left[1-\frac{M_0\omega^2-4D_0^{\alpha\alpha}[\sin^2(k_xa/2)+\sin^2(k_ya/2)]}{C_{0,1}^{\alpha\alpha}}\right], \tag{10}$$

$$Q_N^{\alpha\alpha}=2\left[1-\frac{M_N\omega^2-4D_N^{\alpha\alpha}[\sin^2(k_xa/2)+\sin^2(k_ya/2)]}{C_{N,N-1}^{\alpha\alpha}}\right]. \tag{11}$$

It is obvious that in defining the form of the functions Q_f different types of broken symmetry can be taken into account.

III. CONTINUAL APPROXIMATION AND A PARABOLIC MODEL OF DEFORMATION

In Eqs. (6)–(8) we shall go from the discrete variable f to a continual variable z using the following prescription:

$$\begin{aligned}
&f\to z, \quad N\to L\ ,\\
&U_f^{\alpha}\to U(z), \quad U_1^{\alpha}\to U(a), \quad U_0^{\alpha}\to U(0), \quad U_{N-1}^{\alpha}\to U(L-a), \quad U_N^{\alpha}\to U(L)\ ,\\
&M_f^{\alpha}\to M(z), \quad C_{f,f+1}^{\alpha\alpha}+C_{f,f-1}^{\alpha\alpha}\to 2C(z), \quad C_{0,1}^{\alpha\alpha}\to C(0), \quad C_{N,N-1}^{\alpha\alpha}\to C(L)\ ,\\
&D_f^{\alpha\alpha}\to D(z), \quad U_{f+1}^{\alpha}+U_{f-1}^{\alpha}\to U(z+a)+U(z-a)\approx 2U(z)+a^2\frac{d^2U(z)}{dz^2}\ ,
\end{aligned} \tag{12}$$

where L is the thickness of the crystal in the z direction. Since transitions (12) are a good approximation only in the long-wave range, we should also show that

$$\sin^2\frac{k_xa}{2}+\sin^2\frac{k_ya}{2}\approx\tfrac{1}{4}a^2(k_x^2+k_y^2)=\frac{a^2k^2}{4}\ . \tag{13}$$

In accordance with Eqs. (12) and (13), the difference equations (6)–(8) become

$$\frac{d^2U(z)}{dz^2}+\left[\frac{\omega^2}{a^2}\frac{M(z)}{C(z)}-\frac{D(z)}{C(z)}k^2\right]U(z)=0\ , \tag{14}$$

$$U(a)-2\left[1-\omega^2\frac{M(0)}{C(0)}-\frac{D(0)}{C(0)}a^2k^2\right]U(0)=0\ , \tag{15}$$

$$U(L-a)-2\left[1-\omega^2\frac{M(L)}{C(L)}+\frac{D(L)}{C(L)}a^2k^2\right]U(L)=0\ . \tag{16}$$

For further analysis it is necessary to define the functions $M(z)/C(z)$ and $D(z)/C(z)$, i.e., to choose a model of deformation. We shall assume that all the atoms are identical, i.e., $M(z)=M=\text{const}$. It will also be shown that

$$D(z)=C(z)\ . \tag{17}$$

The last simplifying condition, which can only be reached artificially, will be discussed later. We shall formulate a parabolic model of deformation under the above assumptions. We shall show that the function $M/C(z)$ has the maximal value for $z=L/2$, i.e., $\max M/C(z)=M/C(L/2)\equiv M/C$ and that it decreases symmetrically when z tends to the boundaries of the crystal, i.e.,

$$\frac{M}{C(z)}=\frac{M}{C}-g^2\left[z-\frac{L}{2}\right]^2\ . \tag{18}$$

The values of the strain constant, due to the symmetry assumed, have to be equal at both boundaries, i.e.,

$$C(0)=C(L)=C+C'\ , \tag{19}$$

where the correction C' depends upon the physical conditions on the boundary surfaces. The constant g^2 will be defined from the condition

$$\frac{M}{C(0)}=\frac{M}{C(L)}=\frac{M}{C}-g^2\frac{L^2}{4}=\frac{2M}{C+C'}\ . \tag{20}$$

This condition is chosen in accordance with Eqs. (10) and (11), from where it follows that the ratio M/C is doubled at the boundaries with respect to its bulk values.

From Eq. (20) it follows that

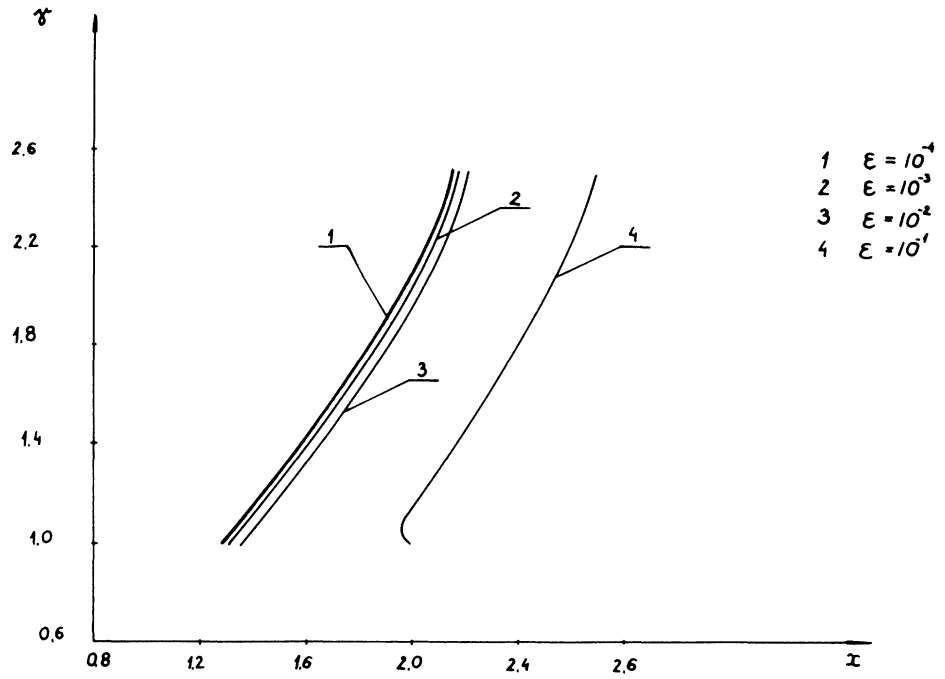

FIG. 1. Dependence of force constant ratio γ on dimensionless wave vector x for various values of relative thickness ϵ.

$$g=\frac{2}{L}\left[\frac{M}{C}\frac{C'-C}{C'+C}\right]^{1/2} . \tag{21}$$

It is clear from (21) that the parabolic dependence (18) is valid only if

$$|C'|>C . \tag{22}$$

It is obvious that condition (22) can be satisfied artificially, let us say, by applying a very high pressure at the boundaries. The condition $D(z)=C(z)$ requires a pressure distribution along the z direction which would achieve such an equality. It is clear that a practical realization of the mentioned pressure distribution is not a simple problem[11,12] and the question remains: is it possible in general. We shall assume that both conditions (17) and (22) are realized in some way and will continue with our calculations.

Substituting (18) into (14)–(16), we obtain the following set of equations:

$$\frac{d^2U(z)}{dz^2}+\left[\frac{M\omega^2}{Ca^2}-k^2-g^2\frac{\omega^2}{a^2}\left[z-\frac{L}{2}\right]^2\right]U(z)=0 , \quad z\in(0,L) , \tag{23}$$

$$U(a)-2\left[1-\omega^2\frac{M}{C'+C}+a^2k^2\right]U(0)=0 , \tag{24}$$

$$U(L-a)-2\left[1-\omega^2\frac{M}{C'+C}+a^2k^2\right]U(L)=0 . \tag{25}$$

After substitution

$$z-\frac{L}{2}=\lambda\xi, \quad \lambda=\frac{aL}{2\omega}\left[\frac{C}{M}\frac{C'+C}{C'-C}\right]^{1/2}, \quad \xi\in\left[-\frac{L}{2\lambda},\frac{L}{2\lambda}\right] \tag{26}$$

Eq. (23) becomes the Hermite-Weber equation

$$\frac{d^2U}{d\xi^2}+\left[\lambda^2\left[\frac{M\omega^2}{Ca^2}-k^2\right]-\xi^2\right]U=0 . \tag{27}$$

In order to avoid enormously high boundary displacements, we shall use the usual condition for an equation of the type (27), i.e.,

$$\lambda^2\left[\frac{M\omega^2}{Ca^2}-k^2\right]=2n+1, \quad n=0,1,2,\ldots , \tag{28}$$

from where we find the phonon frequencies in the considered structures:

$$\omega_n=(2n+1)\frac{a}{L}\left[\frac{C}{M}\frac{C'-C}{C'+C}\right]^{1/2}+\left[(2n+1)^2\frac{a^2C}{L^2M}\frac{C'+C}{C'-C}+\frac{Ca^2k^2}{M}\right]^{1/2} . \tag{29}$$

Taking approximately that $\pm L/2\lambda\rightarrow\pm\infty$ we obtain the following formula for the displacements:

$$U_n(\xi)=\frac{e^{-\xi^2/2}H_n(\xi)}{(2^n n!\pi^{1/2})^{1/2}}, \quad H_n(\xi)=(-1)^n e^{\xi^2}\frac{d^n}{d\xi^n}e^{-\xi^2} . \tag{30}$$

Since the boundary equations (24) and (25) have to be satisfied, the parameters figuring in (29) are not independent. Their dependence will be analyzed here only for the ground-state frequency, i.e., for $n=0$, when formulas (29) and (30) have the form

$$\omega_0=\frac{a}{L}\left[\frac{C}{M}\frac{C'-C}{C'+C}\right]^{1/2}+\left[\frac{a^2C}{L^2M}\frac{C'-C}{C'+C}+\frac{Ca^2k^2}{M}\right]^{1/2}, \tag{31}$$

and

$$U_0(z)=\pi^{-1/4}e^{-[z(L/2)]/2\lambda^2}. \tag{32}$$

After substitution (32), Eqs. (24) and (25) reduce into a unique relation, connecting the characteristic parameters of this theory (we show that $C'>0$):

$$\begin{aligned} &1+x^2-\frac{F^2(x)}{1+\gamma}=\tfrac{1}{2}\exp[(1-\epsilon)F(x)], \\ &x=ak,\quad \gamma=\frac{C'}{C},\quad \epsilon=\frac{a}{L}, \\ &F(x)=\epsilon\left[\frac{\gamma-1}{\gamma+1}\right]^{1/2}+\left[x^2+\epsilon^2\frac{\gamma-1}{\gamma+1}\right]^{1/2}. \end{aligned} \tag{33}$$

In accordance with $|C'|>C$ and $C'>0$ the minimal value of γ is equal to one.

Condition (33) was analyzed numerically for $\epsilon=10^{-4}$, 10^{-3}, 10^{-2}, and 10^{-1}. The results of these calculations are given in Fig. 1. The minimal values of the frequencies ω_0 are given by the formula

$$\min\omega_0^{(i)}=\left[\frac{C}{M}\right]^{1/2}\min F_i(x),\quad i=1,2,3,4 \tag{34}$$

where

$$\begin{aligned} &\min F_1(x)=1.319\,521,\quad \min F_2(x)=1.323\,567, \\ &\min F_3(x)=1.365\,537,\quad \min F_4(x)=1.937\,693. \end{aligned} \tag{35}$$

IV. SUMMARY

From the formula of (35), the following two important conclusions can be drawn.

(a) Only optical phonons can appear in the structure considered.

(b) The minimal energy of the optical phonons increases with decreasing thickness of the film and, consequently, the population of phonons decreases rapidly.

The above conclusions could be of certain importance in the synthesis of high-temperature superconductors, if applied to the metallic structures, since the small phonon population enhances the stability of the superconductive energy gap.[12]

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