Anharmonic perturbation theory for the lattice-dynamic shell model

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We construct a perturbation theory to compute the phonon response function and thermodynamic properties of ionic solids described by an anharmonic shell model.

I. INTRODUCTION

The study of dynamic and thermodynamic properties of a solid beyond the harmonic approximation is of great interest. In particular, the anharmonic effects in the crystal response to neutron scattering are to date well known. This can be qualitatively summarized by saying that a complex anharmonic self-energy modifies each harmonic phonon frequency. The real part leads to a change in the frequency value, while the imaginary part is the reciprocal of the single-particle amplitude for the phonon lifetime. This last effect is observed as a broadening of the peaks in the energy distribution of the scattered neutrons. All these facts have been theoretically understood by using many-body techniques. In particular, diagrammatic expansions for the perturbative evaluation of the renormalized phonon frequencies and the phonon lifetime were used by Maradudin and Fein,[1] for a rigid-ion model.

For the lattice dynamics of ionic crystals the so-called shell model was used with considerable success.[2] In this model, the outer electrons of the ions are represented by a spherically symmetric massless shell of charge. The polarizability effects are thus incorporated in the dipole approximation. This leads to a better description of the observed dynamical properties, as compared with the rigid-ion model.[3] In the harmonic approximation the usual treatment is to eliminate the shell coordinates from the equations of motion by using the adiabatic condition. Therefore, an effective potential for the motion of the cores (nuclei) is given.[2] However, in a general anharmonic situation the shell coordinates cannot be explicitly obtained from the adiabatic condition. Therefore, the formulation of the dynamics and statistical mechanics of the shell model is not trivial. By means of a perturbative method the self-consistent phonon approximation[4] has been recently extended to include calculations with a general anharmonic shell model.[5] However, the method proposed in Ref. 5 is valid only to evaluate the free energy of the system. In a recent work we have obtained the quantum partition function for a general adiabatic shell model by using the path-integral representation.[6]

The aim of this paper is to find a perturbative method to evaluate the phonon response function (susceptibility, Green's functions) and thermodynamic properties for a solid described by an anharmonic shell model. For this purpose we will take as a starting point the partition function obtained in Ref. 6.

II. PRELIMINARIES

The partition function for a general rigid-ion model is:

$$Z=\int_{\text{periodic}}\mathcal{D}u\,\exp\left[-\frac{1}{h}\int_0^{\beta h}d\tau\left[\tfrac{1}{2}M_{ij}\dot{u}^{\,i}\dot{u}^{\,j}+\tfrac{1}{2}D_{ij}u^{i}u^{j}+\Phi_A(u)\right]\right], \tag{2.1}$$

where u^i denotes an ionic displacement whose Cartesian component, cell, and ionic site are summarized in the index i. The quantity $\dot{u}^{\,i}$ is the time derivative with respect to the Euclidean time variable $\tau=it$, and D_{ij} and M_{ij} are, respectively, the dynamic and mass matrices.

The quantity Φ_A is the anharmonic piece of the interacting potential and is given by

$$\Phi_A=\Phi_3+\Phi_4+\cdots, \tag{2.2}$$

where

$$\Phi_n=F_{i_1 i_2\cdots i_n}u^{i_1}u^{i_2}\cdots u^{i_n}. \tag{2.3}$$

By writing the displacement u^i in the basis of eigenvectors A which diagonalizes completely the harmonic part of the potential, the expression (2.1) takes the form

$$Z=\int\mathcal{D}A\,\exp\left[-\frac{1}{h}\left(\sum_{q_1q_2}g^0_{q_1q_2}A_{q_1}A_{q_2}+\sum_{q_1\cdots q_n}F_{q_2q_2\cdots q_n}A_{q_1}A_{q_2}\cdots A_{q_n}\right)\right]. \tag{2.4}$$

In (2.4) we have written the free phonon propagator given by

$$g^0_{q_1q_2}=g^0_q\delta_{q_1q_2}\ , \tag{2.5}$$

where

$$g^0_q\equiv g^0_k(\omega_n)=\frac{1}{-\omega_n^2+\omega_k^2}\ , \tag{2.6}$$

with $\omega_n=(2\pi/\beta h)n$ the Matsubara frequency and ω_k the eigenvalues of the dynamical matrix. The index k corresponds to the set of indices (k,j). The first index runs over the Brillouin zone and the second one is a polarization index. The nth interaction term in the q representation is given by $F_{q_1q_2\cdots q_n}$. By means of these definitions, the diagramatic representation in q space is directly obtained.[1,7]

On the other hand, the anharmonic Green's functions are given by adding diagrams with two external legs. Moreover, it is well known from Dyson's theorem that the correction to the free phonon frequencies $\Delta_{kk'}$ and the inverse $\Gamma_{kk'}$ of the phonon lifetimes are obtained by making

$$\lim_{\epsilon\to 0}\sum_{kk'}(\Omega+i\epsilon)=-\beta h[\Delta_{kk'}(\Omega)-i\Gamma_{kk'}(\Omega)]\ . \tag{2.7}$$

The quantity $\sum_{kk'}(\Omega+i\epsilon)$ is the sum of all the diagrams strongly connected up to a given order, analytically continued to all the complex frequency plane.[7]

In the rigid-ion model the electronic cloud effects are taken into account in the interaction potential between ions. It is known that a suitable way to incorporate the polarizability effects is by treating the electrons as massless shells, with empirical potentials between shells and between cores and shells. The equations of motion to study the lattice dynamic of systems which are described by the shell model, can be obtained from the following Lagrangian:

$$L(\dot u,u,v)=\tfrac{1}{2}\dot u^{\,i}M_{ij}\dot u^{\,j}-\Phi(u,v)\ , \tag{2.8}$$

where v_j is the displacement of the jth spherically symmetric shell and Φ is a general interacting potential.

The Euler-Lagrange equations are

$$M_{ij}\ddot u^{\,j}+\frac{\partial\Phi}{\partial u^{\,i}}=0\ , \tag{2.9a}$$

$$\frac{\partial\Phi}{\partial v_i}=0\ , \tag{2.9b}$$

where this last equation is the adiabatic condition.

The quantum partition function for this model is given by[6]

$$Z=\int_{\substack{\text{periodic}\\ \text{path}}}\mathcal{D}u\ \mathcal{D}v\,\mathrm{Det}[\mathcal{P}]\delta(\chi)\exp\left[-\frac{1}{h}S_E(u,v)\right]\ , \tag{2.10}$$

where the Euclidean action is

$$S_E(u,v)=\int_0^{\beta h}d\tau[\tfrac{1}{2}\dot u^{\,i}M_{ij}\dot u^{\,j}+\Phi(u,v)]\ . \tag{2.11}$$

The expression (2.10) for Z also can be written as

$$Z=\int\mathcal{D}u\ \mathcal{D}v\ \mathcal{D}\lambda\ \mathcal{D}\eta\ \mathcal{D}\eta^\dagger\exp\left[-\frac{1}{h}S'(u,v,\lambda,\eta,\eta^\dagger)\right]\ , \tag{2.12}$$

where the effective action S' is defined by

$$S'(u,v,\lambda,\eta,\eta^\dagger)=S_E(u,v)+\int_0^{\beta h}d\tau[\lambda_i\chi^i+\eta_i^\dagger\mathcal{P}^{ij}\eta_j]\ , \tag{2.13}$$

where

$$\mathcal{P}^{ij}(u,v)=\frac{\partial^2\Phi}{\partial v_i\partial v_j}\ , \tag{2.14}$$

$$\chi^j(u,v)\equiv-\frac{\partial\Phi}{\partial v_j}\ . \tag{2.15}$$

In (2.12) we have used the integral representation for the δ function and the $\det[\mathcal{P}]$ was written as a path integral on the Grassmann variables η.

III. PERTURBATIVE EVALUATION OF THE PHONON RESPONSE FOR THE SHELL MODEL.

With the aim of giving a perturbative method to evaluate the phonon thermodynamic Green's functions we take as a starting point the partition function expression (2.12). The potential $\Phi(u,v)$ will be in general a polynomial in the u^i and v_i variables and therefore the last term on the right-hand side of Eq. (2.13) will also be a polynomial expression in the u, v, λ, η, and $\eta^\dagger$ variables.

If we define the quantity

$$X(i)=\begin{bmatrix}u^i\\ v_i\\ \lambda_i\end{bmatrix}\ , \tag{3.1}$$

the expression (2.13) can be written in terms of it and will contain terms of the form

$$A_{\alpha\beta\cdots\gamma}(i,j,\ldots,k)X^\alpha(i)X^\beta(j)\cdots X^\gamma(k)\ , \tag{3.2}$$

$$B_{\alpha\beta\cdots\gamma}(i,j,\ldots,k,h,p)X^\alpha(i)X^\beta(j)\cdots X^\gamma(k)\eta_h^+\eta_p\ . \tag{3.3}$$

We use the convention of summation on repeated indices for both the Greek and italic ones. The Greek indices run from 1 to 3. In the repeated italic indices, in addition to the summation over sites, an integral from 0 to βh in the continuum variable τ must be understood. We will use this convention whenever the integral on τ is not explicitly written.

In order to construct the matrices A and B appearing in (3.2) and (3.3), the indices of sites must be fixed and the matrix coefficients $A_{\alpha\beta\cdots\gamma}$ and $B_{\alpha\beta\cdots\gamma}$ will be determined in such a way that the development of terms of the form (3.2) and (3.3) allows us to obtain the expression (2.13) for a given potential Φ. In the way the matrix coefficients of the polynomial are constructed, we can see that the Greek and italic indices are not independent.

That is, permutations in the Greek indices imply permutations in the italic ones.

Now we can consider the usual Feynman rules for a given theory. That is to say, the propagator will be given by the harmonic piece of the action and the remaining anharmonic piece will be represented by vertices.[8]

The harmonic piece Φ_0 of the potential Φ can be written as follows:

$$\Phi_0(u,v)=\tfrac{1}{2}R_{ij}u^i u^j+T_i^j u^i v_j+\tfrac{1}{2}S^{ij}v_i v_j \ . \tag{3.4}$$

In this case the expression (2.13) for the effective action S' takes the form

$$S'_0(u,v,\lambda,\eta^\dagger,\eta)=\int_0^{\beta h}(\tfrac{1}{2}M_{ij}\dot{u}^i\dot{u}^j+\tfrac{1}{2}R_{ij}u^i u^j+T_i^j u^i v_j +\tfrac{1}{2}S^{ij}v_i v_j+T_i^j u^i\lambda_j +S^{ij}v_i\lambda_j+S^{ij}\eta_i^\dagger\eta_j)d\tau \ , \tag{3.5}$$

where we have used the following equations:

$$\frac{\partial\Phi_0}{\partial v_m}=T_i^m u^i+S^{im}v_i \ , \tag{3.6}$$

$$\frac{\partial\Phi_0}{\partial v_t\partial v_m}=S^{tm} \ . \tag{3.7}$$

We have used a notation to make evident the lack of symmetry in the coupling constant present in the potential Φ under permutations of the indices. Therefore, we use upper- and lower-site indices for both the displacement fields and the coupling constants.

The above lack of symmetry is due to the distinction between the strength acting on the ith ion when the shell jth is displaced and the strength acting on the jth ion when the ith shell is displaced.

Using the definition (3.1), the expression (3.5) for S'_0 can be written as follows:

$$S'_0=\tfrac{1}{2}X^\alpha(i)[G^{-1}(i,j)]_{\alpha\beta}X^\beta(j)+\eta_i^\dagger S^{ij}\eta_j \ , \tag{3.8}$$

where the matrix G^{-1} whose elements are $[G^{-1}(i,j)]_{\alpha\beta}$ is given by

$$G^{-1}=\begin{bmatrix} g^{-1} & T & T \\ T^+ & S & S \\ T^+ & S & 0 \end{bmatrix} . \tag{3.9}$$

Looking at the expression (3.8) we can see that in this formalism there are two propagators, one for the field X^α and another for the ghost field η. These propagators are given by the matrices $[G(i,j)]^{\alpha\beta}$ and S_{ij}^{-1}, respectively. The inverse of the matrix (3.9) is

$$G=\begin{bmatrix} g & gC^\dagger & 0 \\ Cg & CgC^\dagger & S^{-1} \\ 0 & S^{-1} & -S^{-1} \end{bmatrix} . \tag{3.10}$$

In (3.10) we have called g and C the matrices whose elements are

$$g_{ij}(\tau)=(M_{ij}\partial_\tau^2+D_{ij})^{-1} \ , \tag{3.11}$$

$$C_{ij}=-S_{ik}^{-1}(T^T)_j^k \ , \tag{3.12}$$

where the elements of the matrix D in (3.11) are defined by

$$D_{ij}=R_{ij}-T_i^k S_{kl}^{-1}(T^T)_j^l \ . \tag{3.13}$$

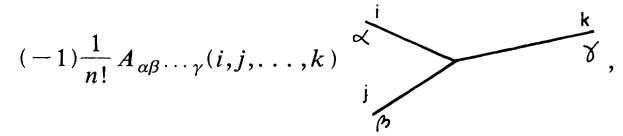
The matrix D is the well-known dynamical matrix in real space. The matrix g is the free-phonon propagator of the effective rigid-ion model. Such propagator is obtained when the adiabatic condition (2.9b) for the potential Φ_0 is used to write v_i as a function of u^i.

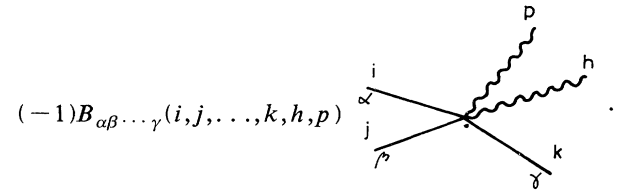
Now we are going to give the Feynman rules in real space for a general anharmonic interacting potential.

(i) Propagators: We associate to the propagator $G_{\alpha\beta}(i,j)$ a line connecting the two points i and j,

We associate to the propagator S_{ij}^{-1} a wavy line connecting the two points i and j,

(ii) Vertices: The generic vertices (of n legs $X^\alpha(i)$) are represented by





(iii) A minus sign must be added to each closed loop built up to η fields, owing to its Grassmannian character.

(iv) Each diagram must be multiplied by the corresponding topological factor.

(v) The summation over all internal indices, Greek and italic, must be carried out.

(vi) The external legs can take only the value $\alpha=1$ corresponding to the first component of the field X^α, which is the unique independent dynamical field.

Therefore the other two components (v and λ) of the field X^α and the ghost field η can contribute only to the internal structure of the diagrams.

Now we are going to illustrate the above considerations with an example which allows us to check our perturbative method. We take a model with the following potential:

$$\Phi(u,v)=\tfrac{1}{2}R_{ij}u^i u^j+T_i^j u^i v_j+\tfrac{1}{2}S^{ij}v_i v_j +\tfrac{1}{2}F_{ij}^k u^i u^j v_k+\tfrac{1}{2}L_i^{jk}u^i v_j v_k \ , \tag{3.14}$$

where R, T, S, F, and L are matrices which give the atomic force constants.

Using the equations

$$\frac{\partial\Phi}{\partial v_m}=T_i^m u^i+S^{im}v_i+\tfrac{1}{2}F_{ij}^m u^i u^j+L_i^{jm}u^i v_j\ , \tag{3.15a}$$

$$\frac{\partial\Phi}{\partial v_l\partial v_m}=s^{lm}+L_i^{lm}u^i\ , \tag{3.15b}$$

and the definition (3.1), the expression (2.13) for the effective action takes the form

$$\begin{aligned}S'=&\tfrac{1}{2}X^\alpha(i)[G^{-1}(i,j)]_{\alpha\beta}X^\beta(j)+\eta_i^\dagger S^{ij}\eta_j\\&+\frac{1}{3!}A_{\alpha\beta\gamma}(i,j,k)X^\alpha(i)X^\beta(j)X^\gamma(k)\\&+B_\alpha(i,j,k)X^\alpha(i)\eta_j^\dagger\eta_k\ .\end{aligned} \tag{3.16}$$

In the present case the only nonvanishing matrix elements are

$$A_{112}(i,j,k)=A_{121}(i,k,j)=A_{211}(k,i,j)=F_{ij}^k\ , \tag{3.17a}$$

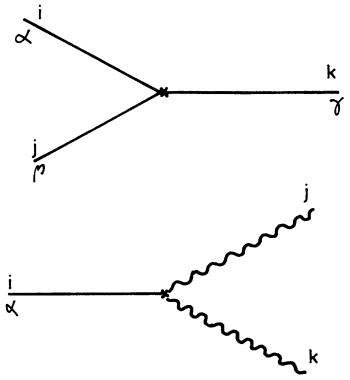
$$A_{113}(i,j,k)=A_{131}(i,k,j)=A_{311}(k,i,j)=F_{kj}^i\ , \tag{3.17b}$$

$$A_{122}(i,j,k)=A_{212}(j,i,k)=A_{221}(j,k,i)=L_i^{jk}\ , \tag{3.17c}$$

$$\begin{aligned}A_{123}(i,j,k)&=A_{132}(i,k,j)=A_{321}(k,j,i)=A_{213}(j,i,k)\\&=A_{312}(k,i,j)=A_{231}(j,k,i)=L_i^{jk}\ ,\end{aligned} \tag{3.17d}$$

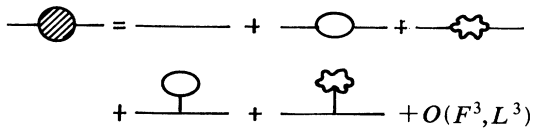
$$B_1(i,j,k)=L_i^{jk}\ . \tag{3.17e}$$

The two vertices corresponding to the interacting potential (3.14) are



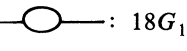
(3.18)

The diagrams with two external legs, containing up to two vertices (second order in the coupling constants) are

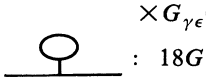


(3.19)

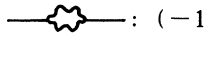
Using the diagramatic rules given above, the analytic expression for each diagram is:



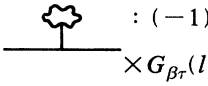
$$\times G_{\gamma\epsilon}(m,q)A_{\delta\epsilon\tau}(p,q,h)G_{\tau 1}(h,j)\ , \tag{3.20a}$$



$$\times G_{\epsilon\tau}(q,t)A_{\delta\epsilon\tau}(l,q,t)G_{\gamma 1}(p,j)\ , \tag{3.20b}$$

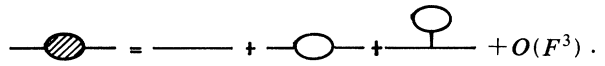


$$\times S_{qt}^{-1}B_\gamma(l,m,t)G_{\gamma 1}(l,j)\ , \tag{3.20c}$$



$$\times G_{\beta\tau}(l,p)G_{\gamma 1}(m,j)B_\tau(p,h,t)S_{ht}^{-1}\ . \tag{3.20d}$$

If we consider (3.14) for a particular case in which $L_i^{jk}=0$, from (3.16) we can see that the ghost and the X^α fields are not interacting. Hence the ghost fields can be integrated out of the path integral and the diagrams (3.20c) and (3.20d) do not appear. Therefore, up to $O(F^3)$ we have



(3.21)

By developing these diagrams the following expression can be written

$$\begin{aligned}&+\tfrac{9}{2}(g_{pl}X_{\langle lth\rangle}g_{ti}g_{hj}X_{\langle ijk\rangle}g_{km})\\&+\tfrac{9}{2}(g_{pl}X_{\langle lth\rangle}g_{hl}X_{\langle ijk\rangle}g_{jk}g_{tn})\\&+\tfrac{3}{2}(g_{pl}X_{\langle lhpj\rangle}g_{hp}g_{jm})\ ,\end{aligned} \tag{3.22}$$

where

$$X_{\langle ijh\rangle}=F_{(ij}^k S_{|kl|}^{-1}T_{h)}^l\ , \tag{3.23a}$$

$$X_{\langle mnij\rangle}=F_{(mn}^t S_{|tl|}^{-1}F_{ij)}^l\ . \tag{3.23b}$$

The parenthesis means that the expression has been symmetrized in all the indices enclosed. The indices enclosed in vertical bars are understood as excluded from the symmetrization. This symmetrization comes naturally from the explicit computation of the terms in the diagrams (3.21).

On the other hand, for this particular potential we are treating, it is possible to define an effective rigid-ion theory. This can be done by writing the field v_i as a function of the u^i through the adiabatic condition (2.9b). Later on, the field v_i thus written must be replaced into the potential. Then we obtain

$$\Phi^{\rm eff}(u)=\Phi_0^{\rm eff}(u)+\Phi_3^{\rm eff}(u)+\Phi_4^{\rm eff}(u)\ , \tag{3.24}$$

where

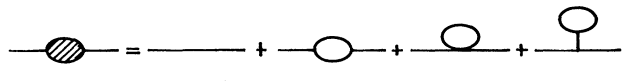
$$\Phi_0^{\rm eff}(u)=\tfrac{1}{2}u^i(R_{ij}-T_i^h S_{hm}^{-1}T_j^m)u^j\ , \tag{3.25a}$$

$$\Phi_a^{\rm eff}(u)=-\tfrac{1}{2}T_i^j S_{jm}^{-1}F_{lp}^m u^t u^l u^p\ , \tag{3.25b}$$

$$\Phi_4^{\rm eff}(u)=-\tfrac{1}{8}F_{lq}^j S_{jm}^{-1}F_{ph}^m u^l u^q u^p u^h\ . \tag{3.25c}$$

Therefore the effective theory contains vertices of three or four legs. From (3.25) we see that only the symmetric part of the factors multiplying the fields u^i must be taken into account.

Now we can compare this last result with the previous one obtained in the framework of the general treatment. To do this we must add diagrams up to $O(F^3)$ which in the last case are



$$+O(F^3)\ . \tag{3.26}$$

The evaluation of the diagrams (3.26) by means of the usual rules of the rigid-ion theory in real space, leads to the same results as those obtained in (3.22).

In summary, we have given the diagrammatic rules for the evaluation of anharmonic properties in a shell model with a generic interacting potential, even if there is no possibility of writing v_i as a function of u^i. In the particular case in which the anharmonicity is such that the effective potential $\Phi(u,v(u))$ can be defined, our results, using the perturbative scheme developed here, are the same as those obtained from the usual anharmonic crystal theory.

For the computation of the phonon thermodynamic Green's functions through a perturbative formalism, it is more convenient to use a representation in a Fourier space. This Fourier representation is obtained by writing the diagrams of the type (3.22) given in a real space, in a representation where g is diagonal. This transformation remains defined by the unitary matrix J which verifies:

$$J^{\dagger}_{qi} g_{ij} J_{jq'} = g^0_q \delta_{qq'}\ , \tag{3.27}$$

where g^0_q is the free-phonon propagator given in (2.6) and in this case ω_k are the eigenvalues of the dynamical matrix of the shell model. Consequently, we must insert unities $J^{\dagger}J$ in the analytic expressions (3.22) of the diagrams. Thus, the first term of the diagrams gives rise to the expression (2.6) for the free propagator g in q space. The other terms give rise to the corresponding expression for the different quantities X in q space.

It is easy to see that the added terms to the harmonic propagators contain anharmonic constants. Therefore, they are corrective to this free propagator.

Having this fact in mind we propose for the shell model the following generalization of the Dyson theorem given in the rigid-ion theory. We assume that the corrections to the frequency and lifetime of the phonon are formally given in the expression (2.7). But we interpret that the quantity $\sum_{kk'}$ is now given by the sum of all the terms obtained from the diagrams strongly connected to the general theory which has the external g removed. We note that diagrams strongly connected in the general theory give rise to expressions which never can be separated into two or more independent pieces by only taking out one g. Finally, as in the rigid-ion theory, the sum of the vacuum-connected diagrams allows us to compute up to a given order the partition function Z and therefore thermodynamical properties can be obtained.

Before concluding this section we wish to remark that our diagrammatic expansion can be simplified by taking into account that the Greek and italic indices are not independent. Therefore the set of italic indices can be eliminated.

Consequently, considering the analytic expression for a given diagram, and after computing the matrix product in the Greek indices, we proceed as follows. (a) First, we order all the terms and consider that two of them are equal when one can be obtained from the other by interchanging the order of the factors. (b) The matrices g appearing in each term of the sum will be interpreted as "propagators," and with the remaining pieces we construct matrices which can be interpreted as "vertices." (c) Subsequently, to reintegrate the indices we proceed as follows: The S^{-1} matrix must be used as a connector between the other matrices. Thus, a summation over the lower indices of this matrix and the upper indices of the remaining matrices must be carried out. (d) Finally, the matrices which represent "vertices" must be symmetrized in all their indices. A summation over the indices of these matrices and the indices of the ("propagators") matrix g of all possible forms must be carried out.

IV. THE PHONON LIFETIME IN THE NONLINEAR POLARIZABLE SHELL MODEL

In this section we show that the phonon lifetime for the polarizability model is infinite at lower perturbative order.

The nonlinear polarizability model describes the dynamical properties of a large variety of ferroelectric materials.[9] This model is characterized by a local anharmonic core-shell interaction which corresponds to the nonlinear polarizability of the anion (i.e., O^{2-}). Such anharmonic coupling treated in the self-consistent phonon approximation accounts for the temperature dependence of the ferroelectric soft mode and other measured properties in several materials.[10] But this approximation does not provide a knowledge of the phonon lifetime.

For convenience, we use $w=v-u$ as a relative displacement coordinate instead of the shell coordinate v. In terms of the u and w coordinates the potential of the model can be written as

$$\Phi(u,w)=\tfrac{1}{2}R_{ij}u^i u^j + T^j_i u^i w_j + \tfrac{1}{2}S^{ij}w_i w_j + \frac{1}{4!}F^{ijkl}w_i w_j w_k w_l\ , \tag{4.1}$$

where, in this case, R, T, and S represent the harmonic interaction between u and w, and F is the anharmonic coupling.

Taking into account the definitions of Sec. III, the action is written,

$$S'=\tfrac{1}{2}X^{\alpha}G^{-1}_{\alpha\beta}X^{\beta}+\eta^{\dagger}S\eta+\frac{1}{4!}A_{\alpha\beta\gamma\delta}X^{\alpha}X^{\beta}X^{\gamma}X^{\delta} + \frac{1}{2!}B_{\alpha\beta}X^{\alpha}X^{\beta}\eta^{\dagger}\eta\ , \tag{4.2}$$

where

$$A_{2222}=A_{3222}=A_{2322}=A_{2232}=A_{2223}=B_{22}=F\ . \tag{4.3}$$

In order to use the simplified diagrammatic rules, all the italic indices were suppressed. The lowest-order diagrams are

 $: \frac{-12}{4!} G_{1\alpha} A_{\alpha\beta\gamma\delta} G_{\beta\gamma} G_{\delta 1} ,$ (4.4)

 $: 2 G_{1\alpha} B_{\alpha\beta} S^{-1} G_{\beta 1} .$ (4.5)

Developing the summation over the Greek indices and by using our rules we obtain the following:

(i) The expression of diagram (4.4) contains two different kinds of terms. One of these has a "vertex" given by

$$X_{(jklm)} = F^{npqr} S^{-1}_{nt} S^{-1}_{ps} S^{-1}_{qi} S^{-1}_{rv} T^{t}_{(j} T^{s}_{k} T^{i}_{l} T^{v}_{m)} . \quad (4.6)$$

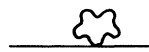
The other one has the following "vertex:"

$$X_{(jk)} = F^{npqr} S^{-1}_{nt} S^{-1}_{pq} S^{-1}_{ri} T^{t}_{(j} T^{i}_{k)} . \quad (4.7)$$

Therefore, we can see that the analytic form of the diagram (4.4) is

 $= -12 \left[\frac{1}{4!} g_{ij} X_{(jlkm)} g_{kl} g_{mq} + \frac{1}{12} g_{ij} X_{(jk)} g_{kq} \right] .$ (4.8)

(ii) The analytic expression for the diagram (4.5) is

 $= 2\left(\frac{1}{2} g_{ij} X_{(jk)} g_{kq}\right) .$ (4.9)

Thus, the perturbative evaluation up to this order leads to the result

 $= -\frac{1}{2} g_{ij} X_{(jklm)} g_{kl} g_{mq} ,$ (4.10)

where the terms without internal propagator g cancel among themselves.

When we consider the Fourier representation of the expression (4.10), we can see that it is equivalent to that corresponding to the tadpole diagram  in a rigid-ion model with quartic anharmonicity given by X. Therefore, there is no contribution to the phonon linewidth up to this perturbative order.

V. CONCLUSIONS

We have constructed a perturbative formalism useful for computing anharmonic properties in a lattice-dynamics shell model. For the case in which an effective potential for the core can be defined, our results are the same as those provided by the rigid-ion theory. Moreover, we have proposed a generalization of the Dyson theorem given in the rigid-ion theory, for the shell model. This generalization allows us to interpret the results of the diagrammatic in terms of corrections of the frequency and lifetime of the phonons.

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