A NUMERICAL RENORMALIZATION GROUP APPROACH FOR AN ELECTRON-PHONON INTERACTION

S. Baher

Department of Physics, Lorestan University, Khoram-Abad, Islamic Republic of Iran

Abstract

A finite chain calculation in terms of Hubbard X-operators is explored by setting up a vibronic Hamiltonian. The model conveniently transformed into a form so that in the case of strong coupling a numerical renormalization group approach is applicable. To test the technique, a one particle Green function is calculated for the model Hamiltonian.

Introduction

Different types of systems in which localized electronic states can be strongly coupled to vibronic modes (phonon) which may occur at defects or impurities in insulators or certain molecules. Examples of this type of problems are Jahn-Teller Systems with degenerate electronic states or mixed-valence molecules. In these systems quantum mechanical tunnelling (dynamic effects) between equivalent configurations may occur in which the spectrum of vibronic levels is calculated. The Hamiltonian of these systems is expressed in matrix form, using products of suitable electron-phonon states as a basis.

The matrix elements subsequently diagonalized numerically, gives the vibronic levels. These types of calculations become too complex and the matrices become too large for numerical techniques.

Here, an iteration scheme which makes use of the numerical renormalization group approach, originally developed by Wilson [1,2] for the Kondo [3,4] problem, is used.

The strong coupling constant of electron-phonon interaction is considered first and then put in a form, so that the rest of the modes can be treated perturbationally. In applying this method to multi-mode electron-phonon systems, phonon modes are coupled in a chain-like fashion/cluster, so that they can be treated iteratively. The phonon modes are arranged so that the first mode is the only one which is vibronically coupled to the multi-level/impurity system in a cluster model [10], Figure 1.

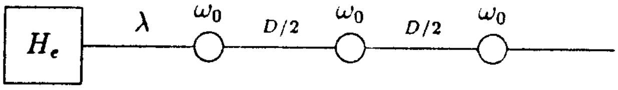

Figure 1. Electronic part, H_e, is coupled to phonon modes through a coupling constant λ.

To realize the scheme, a set of orthogonal basis states is generated using the Lanczos algorithm. The Lanczos method is a procedure used for putting any Hamiltonian matrix form into tridiagonalization form [5]. The method works by making a succesive choice of basis vectors by operating with the matrix on any suitable vector. Here the initial normalized basis vector is X_1, and states H^nX_1, $n=1,2,3,\ldots$ are generated, where H is the original matrix.

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Therefore, X_1 is selected to be the phonon state which is linearly coupled to the electronic system. The Hamiltonian composing electronic system, coupled with the modes, is set up in matrix form using uncoupled electron and phonon states as a basis. The matrix is in block form, with blocks of elements corresponding to a particular number of phonons excited and non-zero off-diagonal blocks linking n-phonon states with $(n+1)$ phonon states.

This matrix must be truncated, and it is usually truncated so that it includes blocks of some specific number of phonon excitations.

The Model Hamiltonian

The theory is developed for a model Hamiltonian in the form:

$$H = H_e + H_p + H_i \tag{1}$$

where H_e is a purely electronic part, H_p describes the vibronic modes in the harmonic approximation and H represents the electron-phonon interaction.

The model Hamiltonian (1) can be expressed in terms of standard basis operators, (Hubbard X-operators), [6,7,8]. The standard operators are those that create and annihilate states of the non-interacting model. They are defined by ${}_{ns}X_{n's'} = |ns\rangle\langle n's'|$, where $|ns\rangle$ are the eigenstates of the uncoupled Hamiltonian. By making use of the above operators, (1) becomes [9,10]:

$$H = \sum_{ns} E_{ns\ ns} X_{ns} + \sum_{k} \omega_k b_k^+ b_k + \sum_{\substack{k \\ ns,\, n's'}} \lambda_k^{ns,\, n's'} (b_k^+ + b_k)_{ns} X_{n's'} \tag{2}$$

where b_k^+, b_k are creation and annihilation operators for the phonon of frequency ω_k, and $\lambda_k^{ns,\, n's'}$ is the matrix element of H_i between states $|ns\rangle$ and $|n's'\rangle$ divided by $\sqrt{2\omega_k}$.

Due to exploring an iterative scheme, a canonical transformation is used, so that in the new representation, the electronic part is coupled in a single mode with creation and annihilation operators B_1^+, B_1 where

$$\lambda B_1^+ = \sum_k \lambda_k b_k^+ \quad , \quad B_1^+ = \sum \tilde{\lambda}_k\ b_k^+ \tag{3}$$

$$\lambda = \sqrt{\sum_k \lambda_k^2} \quad , \quad \tilde{\lambda}_k = \lambda / \lambda_k \tag{4}$$

In the new representation, H_p is no longer diagonal, but takes the form $\sum_{ss'} D_{ss'}\, B_{s'}^+$, $s > 1$, where the orthogonal transformation to the new phonon states is denoted by:

$$B_s = \sum_k \alpha_{ks} b_k \tag{5}$$

$$\alpha_{ks} = \tilde{\lambda}_{k\,s}$$

with α_{ks} real.

A column vector $|s\rangle = (\alpha_{ks})$, where k denotes the components of the vector is introduced. The commutation relation reads:

$$[B_s^+, B_s] = \sum_{k k'} \alpha_{k\,s} \alpha_{k's'}\ [b_k^+, b_k] = \delta_{ss'} \tag{6}$$

The diagonal term $\sum_k \omega_k b_k^+ b_k$ transforms to a non-diagonal form as follows:

$$\sum_k \omega_k b_k^+ b_k = \sum_{ss'} \Big(\sum_k \omega_k\ \alpha_{ks}\ \alpha_{ks'}\Big)\, B_s^+ B_{s'} = \sum_{ss'} D_{ss'} B_s^+\ B_{s'} \tag{7}$$

where the matrix element $D_{ss'}$ is given by

$$D_{ss'} = \sum_k \omega_k\ \alpha_{ks}\ \alpha_{ks'}' \tag{8}$$

The form of the proposed iterative scheme for the model Hamiltonian is illustrated in Figure 2, with diagonal elemenls D_{ss} and off-diagonal ones $D_{ss'}$, $s \neq s'$.

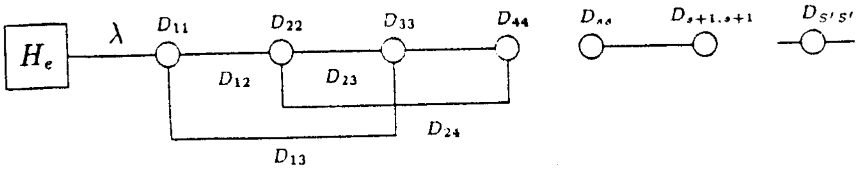

Figure 2. Diagramatic representation of the chain of phonon coupled to multi-level electronic state

The Phonon Basis States

In the following calculations based on Lanczos algorithm, one can show that the α_{ks}'s can be chosen so that only the nearest neighbour matrix elements $D_{ss'}$, $s' = s+1$ need to be taken into account.

This method works by making a successive choice of basis vector in such a way as to produce a transform matrix that is tri-diagonal.

A complete set of orthogonal basis state: $\{|1>, |2>, \ldots |n>, \ldots\}$ can be generated where the first vector $|1>$ is defined as follows:

$$|1> = B^{+}|0> = \sum_{k} \tilde{\lambda}_k \, | \, k > \qquad (9)$$

with $|k> = b_k^{+}|0>$, where $|0>$ is the vacuum of H_p.

The vector $|1>$ is equal to $(\lambda_k) = (\alpha_{k1})$ corresponding to frequency D_{11}.

$$D_{11} = < 1|H_p|1 > = \sum_{k} \omega_k \tilde{\lambda}_k^2 \qquad (10)$$

The second vector $|2>$ is chosen to be orthogonal to $|1>$. The coupling constant D_{12} and D_{22} are calculated as follows:

$$D_{12} = < 1| \, H_p \, | \, 2 > = \sqrt{\left(\sum_{k} \omega_k^2 \, \tilde{\lambda}_k^2 - D_{11}^2\right)} \qquad (11)$$

$$D_{22} = < 2 \, | \, H_p \, | \, 2 > = \frac{D_3 - 2D_{11}D_2 + D_2^3}{D_2 - D_{11}^2} \qquad (12)$$

where $D_3 = <1|H_p^3|1>$, ancl $D_2 = <1|H_p^2|1>$. The procedure is continued to obtain the general form of vectors which corresponds to $|n-1>$, $|n>$ and $|n+1>$ modes along the chain as shown in Figure 3.

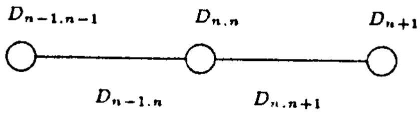

Figure 3. The n-1, n and n+1 terms along the cluster

The $(n+1)^{th}$ phonon state can be written as

$$|n+1> = D_{n,n+1}^{-1} \, [\hat{D}|n > - \tilde{D}_{n,n} \, |n > - D_{n-1,n} \, |n-1> \, | \qquad (13)$$

where $\tilde{D} = H_p$.

The general form of H_p on the basis of the new representation with only the nearest neighbour modes coupled to each other may be written as

$$. \, H_p = \sum_{n=1} D_{n\,n} B_n^{+} B_n + \sum_{n=2} D_{n-1,\,n} \, (B_n^{+} B_{n-1} + B_{n-1}^{+} B_n) \qquad (14)$$

For $\lambda=0$, one has $H_0 = H_e + H_p$ with eigenstates $|n\gamma>$, where γ labels the electronic part and n represents the phonon excitation number.

Application in a Two-Level System

The iterative scheme is applied to a two-level electronic system coupled to phonons. To perform the scheme, first we take into account the first mode of frequency D_{11}, and diagonalize the Hamiltonian matrix H plus the first phonon mode along the chain which is directly coupled to it. Secondly, the diagonalized states of the first stage are used as a basis to take in account the next mode along the chain. Thiss then, is diagonalized. A new Hamiltonian is constructed by using these, to include the next mode along the chain and so on. As an approximation in calculating the lower energy states, the basis is truncated and only a limited number of states is retained in each stage. The model Hamiltonian is given by:

$$\begin{aligned} H = \sum_{i=a,b} E_i c_i^{+} c_i + t(c_a^{+} c_b + c_b^{+} c_a) + \sum \lambda_k \, (b_k^{+} + b_k) \\ (c_a^{+} c_a - c_b^{+} c_b) + \sum_{k} \omega_k \, b_k^{+} b_k \end{aligned} \qquad (15)$$

The creation and annihilation operators c_a^{+}, c_a and c_b^{+}, c_b correspond to the electronic states at sites **a** and **b**, respectively, with an overlap matrix element, t.

To write the Hamiltonian in a slightly different form we introduce bonding and anti-bonding operators, namely,

$$C_{1,0} = 2^{-1/2} \, (c_a \pm c_b), \quad \begin{matrix} + \longrightarrow 1 \\ - \longrightarrow 0 \end{matrix}$$

In terms of these operators we get

$$\begin{aligned} H = (E_0 + t) \, C_1^{+} C_1 + (E_0 - t) \, C_0^{+} C_0 \\ + \sum_{n} \lambda_k \, (b_k^{+} + b_k) \, (C_1^{+} C_0 + C_0^{+} C_1) + \sum_{k} \omega_k \, b_k^{+} b_k \end{aligned} \qquad (16)$$

On the new basis states we have

$$H = (E_0 + t) \, C_1^{+} C_1 + (E_0 - t) \, C_0^{+} C_0 + \sum_{n=1} D_{n\,n} \, B_n^{+} B_n$$

$$+\sum_{n=1} D_{n+1,n}(B_{n+1}^{+}B_n + B_n^{+}B_{n+1}) + \lambda(B_1^{+} + B_1)(C_1^{+}C_0 + C_0^{+}C_1) \qquad (17)$$

We keep the first mode and ignore the rest to diagonalize the Hamiltonian H_1, corresponding to the first stage of our iterative scheme (Fig. 4).

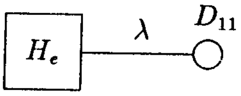

Figure 4. The first mode coupled to electronic system

Then we may write,

$$\begin{aligned} H_1 = {} & (E_0+t)\, C_1^{+}C_1 + (E_0 - t)\, C_0^{+}C_0 \\ & + D_{11}B_1^{+}B_1 + \lambda(B_1^{+} + B_1)(C_1^{+}C_1 + C_0^{+}C_1) \end{aligned} \qquad (18)$$

In terms of Hubbard X-operators we can write,

$$\begin{aligned} H_1 = {} & (E_0+t)\, {}_0X_0 + (E_0 - t)_1 X_1 \\ & + \lambda(B_1^{+} + B_1)({}_1X_0 + {}_0X_1) + D_{11}B_1^{+}B_1 \end{aligned} \qquad (19)$$

For $E_0 = 0$ and in the case of, $\lambda = 0$ by considering the first phonon mode of the chain, we get

$$H_1 = t_0\, {}_0X_0 + t_1\, {}_1X_1 + D_{11}B_1^{+}B_1 \qquad (20)$$

where $t_0 = t$ and $t_1 = -t$.

The eigenstates of diagonal part are given by

$$|n\gamma\rangle = \sqrt{n!}\,(B_1^{+})^n\,|0\rangle\,|\gamma\rangle,\ n = 0,1,2,3,\ldots,\gamma = 0,1 \qquad (21)$$

In this notation $\gamma = 1$ denotes the (+) parity, similarly to a spin-up system, also, $\gamma = 0$, denotes the (–) parity which bears similarity to the spin-down system. The set of basis vectors $\{|n\gamma\rangle, n \in N, \gamma = 0,1\}$ is broken down into two subsets belonging to H_0 subspare

$$\begin{aligned} S_1 &= \{|n\gamma \geq, \gamma = +, \text{for } n \text{ even}, \gamma = -, \text{for } n \text{ odd}\} \\ S_2 &= \{|n\gamma \geq, \gamma = -, \text{for } n \text{ even}, \gamma = +, \text{for } n \text{ odd}\} \end{aligned} \qquad (22)$$

The subsets S_1 and S_2 determine two blocks, the first of which corresponds to

$$H_0^{(1)} = (-1)^n t + nD_{11} \qquad (23)$$

and the second,

$$H_0^{(2)} = -(-1)^n t + nD_{11} \qquad (24)$$

where n denotes the number of excitation phonons. $H_0^{(1)}$ and $H_0^{(2)}$ are related to diagonal elements of (+) and (–) parity blocks, respectively.

The Effective Hamiltonian

In the case of $\lambda \neq 0$, using $|r\alpha\rangle$ to represent the basis states for the first block of H, we may write $|r\alpha\rangle$ as a linear combination of diagonal states (i.e. S_1):

$$|r\alpha\rangle = \sum_{n\gamma} q_{n\gamma}^{r\alpha}\, |n\gamma\rangle \qquad (25)$$

with r representing the phonen excitation number and α denoting the parity of states.

Also, for the negative block, $|s\beta\rangle$ can be written as a linear combination of S_2:

$$|s\beta\rangle = \sum_{n'\gamma'} q_{n'\gamma'}^{s\beta}\, |n'\gamma'\rangle \qquad (26)$$

The matrix elements of H_0 can be established with respect to the above basis

$$H_{nn',\gamma\gamma'} = \langle n\gamma|H_1|n'\gamma'\rangle \qquad (27)$$

with H_1 which is taken from Equation (20).

Here H_1 decomposed into two blocks corresponding to the states $|r\alpha\rangle$ and $|s\beta\rangle$.

The next stage in this process is to numerically diagonalize H_1.

A mode is added to the chain in the next stage, causing expansion of the matrix size, so that we can write

$$H_2 = \widetilde{H}_1 + H_{12} = \sum_{i=0,1} \tilde{t}_i\, {}_iX_i + D_{22} + B_2^{+}B_2 + D_{12}(B_2^{+}B_1 + B_1^{+}B_2) \qquad (28)$$

where $\tilde{t}_i$ stands for the diagonalized form of the preceding part. The eigenstates for this system, namely $|r'\alpha'\rangle$ and $|s'\beta'\rangle$ can be expressed as a linear combination of $|r\alpha\rangle$ and $|s\beta\rangle$.

$$|r'\alpha'\rangle = \sum_{r\alpha, n_2} q_{r\alpha, n_2}^{r'\alpha'}\, |r\alpha, n_2\rangle$$

$$|s'\beta'\rangle = \sum_{s\beta, n_2'} q_{s'\beta'}^{s\beta, n_2'}\, |s\beta, n_2'\rangle \qquad (29)$$

Then the bosonic operators B_1^{+} and B_1 are written in terms of ${}_pX_q$-operators.

$$B_1^+ = \sum_{r\alpha,\, s\beta} \left(g_{r\alpha}^{r\alpha,\, s\beta}\ {}_{r\alpha}X_{s\beta} + \tilde{g}^{s\beta,\, r\alpha}\ {}_{s\beta}X_{r\alpha}\right) \tag{30}$$

where $g^{r\alpha,\, s\beta}$ and $\tilde{g}^{s\beta,\, r'\alpha}$ are real.

Now, H_{12} is evaluated in terms of X-operators. Then the diagonalized form of H_2 for the basis $|r'\alpha'|>$ and $|s'\beta'|$ is calculated.

The matrix elements of the H_2 for each block are constructed such that the alternating states $|r\alpha>$ and $|s\beta>$ are replaced one after the other, each time adding a phonon of the second type.

The matrix elements of H_2 are calculated as

$$\begin{aligned}
&\langle r_1\alpha_1,\ n_2|H_2|\ s_1\beta_1,\ n_2'\rangle = \\
&\langle r_1\alpha_1,\ n_2|\tilde{H}_1 + D_{12},\ (B_1^+B_2 + B_2^+B_1)\ |s_1\beta_1,\ n_2'\rangle \\
&= \tilde{H}_1 \delta_{n_2, n_2'}\ \delta_{r_1\alpha_1,\, s_1\beta_1} \\
&+ D_{12} \sum_{r\alpha,\, s\beta} \left[g^{r\alpha,\, s\beta}\, (n_2+1)^{1/2}\, \delta_{n_2,\, n_2'-1}\ \tilde{g}^{s\beta,\, r\alpha} (n_2)^{1/2} \delta_{n_2,\, n_2'+1}\right]
\end{aligned} \tag{31}$$

For the third mode along the chain the Hamiltonian is written as

$$H_3 = \tilde{H}_2 + D_{33}B_3^+B_3 + D_{23}(B_2^+B_3 + B_3^+B_2) \tag{32}$$

Having expressed H_2 analytically, its Hamiltonian is numerically diagonalized.

The matrix elements of H_3 are worked out in the same method as before;

$$|r''\alpha''> = \sum_{r'\alpha',\, n_3} q_{r'\alpha',\, n_3}^{n''\alpha''}\ |\ r'\alpha',\ n_3>$$

$$|s''\beta''> = \sum_{s'\beta',\, n_3'} \tilde{q}_{s'\beta',\, n_3'}^{s''\beta''}\ |\ s'\beta',\ n_3'> \tag{33}$$

The whole process then continues, each time adding a new mode to the chain which increases the size of the matrix.

One Particle Green Function

The optical transitions are proportionate to the imaginary part of the Fourier transform of response function [11,12,13]:

$$G(t) = -i < 0|TX(t)X(0)|0 > \tag{34}$$

where t = time, $|0>$ = ground state, T= time ordering operator.

A single particle Green function corresponding with a two-level system coupled to a single phonon mode may be written as:

$$G(t) = << {}_0X_1(t):\ {}_1X_0(0) >> \tag{35}$$

where

$$\begin{aligned}
{}_1X_0 = \sum_{r\alpha,\, s\beta} \{ &\left(\sum_{n\ even} q_{n+}^{*r\alpha}\ q_{n-}^{*r\alpha}\right) {}_{r\alpha}X_{s\beta} \\
+ &\left(\sum_{n\ odd} q_{n+}^{*r\beta}\ q_{n-}^{*r\alpha}\right) {}_{s\beta}X_{r\alpha}\}
\end{aligned} \tag{36}$$

$$\begin{aligned}
&= \sum_{r\alpha,\, s\beta} \left(u^{r\alpha,\, s\beta}\ {}_{r\alpha}X_{s\beta} + \tilde{u}^{s\beta,\, r\alpha}\ {}_{s\beta}X_{r\alpha}\right) \\
&u^{r\alpha,\, s\beta} = \sum_{n,\, \gamma\gamma'} q_{n\gamma}^{r\alpha}\, q_{n\gamma'}^{s\beta}
\end{aligned} \tag{37}$$

with correspondence between γ, γ' and n, n' as:

$$a) \begin{cases} \gamma = - \rightarrow n \text{ even} \\ \gamma' = + \rightarrow n' \text{ even} \end{cases}, \qquad b) \begin{cases} \gamma' = - \rightarrow n' \text{ odd} \\ \gamma = + \rightarrow n \text{ odd} \end{cases}$$

$$\begin{aligned}
G_{01}(\omega + i\delta) &= \frac{1}{2\pi}\ \frac{[{}_0X_0 - {}_1X_1]}{[\omega - (t_1 - t_0) + i\delta]} \\
&= \sum_{r\alpha,\, s\beta} \{ \frac{\langle {}_{s\beta}X_{s\beta}\rangle\, |u^{r\alpha,\, s\beta}|^2}{[\omega - (t_{r\alpha} - t_{s\beta}) + i\delta]} \\
&+ \frac{\langle {}_{r\alpha}X_{r\alpha}\rangle\, |\tilde{u}^{s\beta,\, r\alpha}|^2}{[\omega - (t_{s\beta} - t_{r\alpha}) + i\delta]} \}
\end{aligned} \tag{38}$$

with numerically measurable variables $u^{r\alpha,\, s\beta}$, $\tilde{u}^{s\beta,\, r\alpha}$, $\langle {}_{r\alpha}X_{r\alpha}\rangle$ and $\langle {}_{s\beta}X_{s\beta}\rangle$ where

$$\langle {}_{r\alpha}X_{r\alpha}\rangle = \frac{Tr\ {}_{r\alpha}X_{r\alpha}\, e^{-\beta H_1}}{T_r\ e^{-\beta H_1}}\ \} \tag{39}$$

The spectral density of states may be calculated as follows [14]

$$\rho(\omega) = \frac{-1}{\pi}\, Im\ G(\omega + i\delta)$$

$$= \frac{1}{2} \sum_{r\alpha, s\beta} [< {}_{s\beta}X_{s\beta} > |U^{r\alpha, s\beta}|^2 \, \delta[\omega-(t_{sb} - t_{r\alpha})] +$$

$$< {}_{r\alpha}X_{r\alpha} > | U^{r\beta, ra}|^2 \, \delta[\omega - (t_{r\alpha} - t_{s\beta})] \qquad (40)$$

The function $\rho(\omega)$ represents a number of sharp lines which are spread over the range of energy at positions $\Delta E = |t_{r\alpha} - t_{s\beta}|$.

The strength of lines is calculated numerically from $U^{r\alpha, s\beta}$, $U^{s\beta, r\alpha}$, $< {}_{r\alpha}X_{r\alpha} >$ and $< {}_{s\beta}X_{s\beta} >$. The presence of the coupling λ in Equations (19,28) as well as the temperature dependence of some measurable quantities such as $\langle {}_{r\alpha}X_{r\alpha} \rangle$ and $\langle {}_{s\beta}X_{s\beta} \rangle$ affect the strength of the spectral density of states.

Taking into account the second mode along the chain in this diagonalizating process, new variables should be evaluated.

To compare this procedure with those of alternative methods of calculation, the reference [11] is introduced.

Conclusion

In summary an interation scheme which makes use of the numerical renormalization group approach has been used to calculate the spectrum of vibronic levels due to dynamic effects which occured in certain molecules or impurities in insulators.

The Hamiltonian of these systems is expressed in matrix form using products of suitable electron-phonon states as a basis.

In applying the method to multi-mode electron-phonon systems, phonon modes are coupled in a chain-like fashion. Then, a set of orthogonal basis states is generated using lanczos algorithm.

The matrix form of the Hamiltonian is expressed in block form with blocks of elements corresponding to a particular number of phonon excited and non-zero, off-diagonal blocks linking n-phonon states with $(n+1)$ phonon states. Since the matrix elements are subsequently diagonalized numerically to give the vibronic levels, the matrices becomes too large for numerical techniques.

Therefore, for the calculations based on lanczos algorithm, only the nearest neighbour matrix elements along the chain need to be taken into account.

The iterative scheme is then applied to a two-level electronic system coupled to phonons. In performing the scheme the first mode is taken into account, followed by diagonalizing of the Hamiltonian matrix, plus the first phonon mode along the chain which is directly coupled to it. The diagonalized states of the first stage are used as a basis to take into account the next mode along the chain. This; then, is diagonalized. This diagonalization process is continued by considering a new mode along the chain each time. As an approximation in calculating the lower energy levels, the basis is truncated and only a limited number of states is retained.

It is convenient to introduce bonding and antibonding operators, as well as, Hubbard X-operators. Then the Hamiltonian is written in a slightly different form.

Also to obtain a set of basis vectors, which determines two blocks belonging to an uncoupled Hamiltonian, the coupling constant is first set to be zero.

In the case of a non-zero coupling constant, an effective Hamiltonian is calculated, taking new basis states for the blocks as linear combinations of the diagonal subsets. The matrix elements are established with respect to the above basis. The diagonalizing process is used in each stage when a mode is added to the chain.

Finally, to calculate the spectral density of states, a single particle Green function corresponding to a two-level system, coupled to a single mode, is worked out. A number of sharp lines which are spread over the range of energy represent the spectral density of states and the strength of lines is affected by the coupling constant as well as temperature dependence of some measurable quantities.

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