Perturbative expansion using variational phonon basis for Holstein model

Jayita Chatterjee, A. N. Das and P. Choudhury

1 Saha Institute of Nuclear Physics
1/AF Bidhannagar, Calcutta 700 064, India
2 Central Glass and Ceramic Research Institute
196 Raja Subodh Chandra Mullick Road,
Calcutta 700 032, India

A simple variational displacement phonon basis, obtained through the modified Lang-Firsov (MLF) transformation, is proposed to study the Holstein model. This phonon basis contains only one variational parameter, but capable of describing lattice distortions at distant sites from the charge carrier. Perturbation method based on this MLF basis is employed to calculate the single-electron ground-state energy and the dispersion of the polaronic band. The ground-state \((k=0)\) energy obtained up to the second-order perturbation within this approach agrees well with the available numerical results for the entire range of coupling strength.

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degeneracy is lifted by considering the first-order energy
since the electron can occupy any one of the
phonon averaging (ZPA) (i.e., from
many variational parameters. Previously we \[9\] observed
state corresponds to
the total number of phonons in that state. The ground
H unperturbed energy of the state, described in Eq. (3), is

\[ H_0 = \sum_{i} (b_{i}^{\dagger} b_{i} + \frac{\lambda}{2} b_{i}^{\dagger} b_{i} - \delta_{i} b_{i}^{\dagger} b_{i}) \]

where

\[ E_{k=0} = \epsilon_p - 2t_e \] (equivalent to the ground-state energy
obtained by ZPA of \( \tilde{H} \)) with respect to the variational parameters
\( \lambda_1, \lambda_2, ..., \lambda_l \) gives the general recursion relation between the

\[ \frac{\lambda_{l-1}}{\lambda_l} = \frac{\lambda_{l-2} - 2\lambda_{l-1} + \lambda_l}{\lambda_{l-1} - 2\lambda_l + \lambda_{l+1}}. \] (5)

which is satisfied by a simple choice for the parameters
\[ \frac{\lambda_1}{\lambda_0} = \frac{\lambda_2}{\lambda_1} = ... = \frac{\lambda_l}{\lambda_{l-1}} = ... = r. \] (6)

A sum rule is also followed by \( \lambda_i \)'s; for one-dimensional chain this is given by

\[ \lambda_0 + 2\lambda_1 + 2\lambda_2 + ... + 2\lambda_l + ... = g. \] (7)

Eq. (7) is a consequence of the fact that the in-phase phonon mode (zero momentum) being a sum of lattice
displacement operators at different sites \( \left( \sum_{i} (b_{i}^{\dagger} b_{i}) \right) \) couples only with the total number of electrons, which
is a constant of motion. Eqs. (6) and (7) allow one to
consider lattice distortions at all sites of the
infinite chain, contains only one variational parameter.

The polaron self-energy may then be written in terms of \( r \) as \( \epsilon_p = \epsilon - \omega g r (1-r) (1+r^2) \)
and the effective hopping \( t_e = t \exp(-r^2x^2) \) where \( x = \frac{1-r}{1+r}. \)

The expression for the off-diagonal matrix element be-
 tween the ground state and an excited state is given by

\[ H_{l(n),G} = \langle \psi_{l} | H_0 | \psi_{G}^{(0)} \rangle \]

\[ = -t_e y^n \sqrt{N} \left[ \frac{(-1)^{n_1} r^4 + (-1)^{n_R} r^B}{1 + r^2} \right] \]

\[ + \frac{g \omega}{\sqrt{N}} |(1-x)\delta_{n_1,1} - x[r(\delta_{n_1+1,1} + \delta_{n_2+1,1}) + r^2(\delta_{n_2+1,1} + \delta_{n_3+1,1})] | \] (8)

where \( g = gx(1-r), \)
\( n_L = n_{l-1} + n_{l-2} + n_{l-3} + ... \)
\( n_R = n_{l+1} + n_{l+2} + n_{l+3} + ... \)
\( A = n_{l+1} + n_{l+2} + 2(n_{l+2} + n_{l-3}) + 3(n_{l+3} + n_{l-4}) + ... \)
\( B = n_{l-1} + n_{l+2} + 2(n_{l-2} + n_{l+3}) + 3(n_{l-3} + n_{l+4}) + ... \) (9)

The second-order correction to the ground-state energy is given by

\[ E_0^{(2)} = -2g^2 \frac{\omega}{\omega} \left[ \frac{\omega r - t_e (1-r)^2}{(3+r)(1+r)^2} \right] \]

\[ \frac{3}{3+r} \]

\[ = \frac{1}{N} \sum_{n_1} \sum_{n_0 \geq 2} \frac{\epsilon_n^2 + \epsilon_n B}{(n_0 \omega)} \] (10)
and the first-order correction to the ground-state wave function is obtained as,

\[ |\psi_G^{(1)}\rangle = -\frac{1}{\omega\sqrt{N}} \sum_{l=1}^{N} c_l^r |0\rangle_e \{ \omega r - t_e (1 - r)^2 \} \]

\[ \sum_{m} 2g \frac{\delta_{m,t}}{1 + r} - \sum_{p} gxxr^{p-1} \delta_{m,(t+p)} |00..0 1m 0..\rangle_{ph} \]

\[ - \frac{1}{\omega_0} \sum_{l=1}^{N} c_l^r |0\rangle_e \sum_{n_T \geq 2} \frac{H_l(n_l,G)_{n_T}}{n_1 n_2 ... n_N} |n_1 n_2 ... n_N\rangle_{ph} \]

Now one has to make a proper choice of \( r \). In addition to Eq. (6) the minimization of \( E_{k=0} \) yields a condition \( \omega r = t_e (1 - r)^2 \), which gives

\[ r = \frac{(2t_e + \omega) - \sqrt{4t_e \omega + \omega^2}}{2t_e} \]

It is interesting to note that for this choice of \( r \) the off-diagonal matrix element between the ground state and any excited state with a single phonon (\( n_T = 1 \)) becomes zero and this is consistent with our previous studies [7]. Correspondingly, the first term in Eq. (10) as well as in Eq. (11) vanishes.

\[ m_{G}^{(1)} = \frac{m_0}{m_e} - \frac{1}{2} \sum \delta_{k} E(k)_{0} = \frac{1}{2} \sum \frac{\delta_{k} E(k)_{0}}{m_0} \]

\[ m_{G}^{(2)} = \frac{m_0}{m_e} - \frac{1}{2} \sum \delta_{k} E(k)_{0} - \frac{1}{2} \sum \delta_{k} E(k)_{0} \]

\[ m_{G}^{(3)} = \frac{m_0}{m_e} - \frac{1}{2} \sum \delta_{k} E(k)_{0} - \frac{1}{2} \sum \delta_{k} E(k)_{0} - \frac{1}{2} \sum \delta_{k} E(k)_{0} \]

For real materials of interest the Hamiltonian may involve other interactions in addition to the Holstein-type e-ph interaction and the perturbation calculations for such complicated systems may not be possible. The common practice in such cases is to make LF or MLF transformation followed by ZPA. It may be mentioned that ZPA is equivalent to the zeroth (first) order of perturbation as far as the wave function (energy) is concerned. The success of the ZPA depends on the choice of the phonon basis. One of the advantages of the MLF phonon basis over LF basis is that the second-order perturbation correction \( E_0^{(2)} \) within MLF basis is much smaller than that within LF basis except in the strong-coupling regime where both the bases become equivalent. For instance, for \( \omega = t = 1 \), \( E_0^{(2)}(MLF) = -0.01746 \) and \( -0.1849 \) while \( E_0^{(2)}(LF) = -1.3538 \) and \( -0.4801 \) for \( g = 1.0 \) and 1.7, respectively.

A comparison of the effective mass of the polaron obtained within the MLF and LF with ZPA may be helpful. We have calculated the effective mass of the polaron using the standard formula \( m_e^* = \frac{1}{2} \sum \frac{\delta_{k} E(k)}{m_0} \) within the ZPA of LF and MLF Hamiltonian. These results along with the exact numerical result of Ref. [3] are presented in Fig. 1. The proximity of the MLF results with the exact one indicates that the MLF basis is a better choice.
In Fig. 2 we have shown the ground-state energy calculated up to the second-order perturbation correction within the LF (corresponds to \( r = 0 \)) and the MLF phonon basis. Energies calculated from the weak coupling (self-consistent Migdal approximation) and strong coupling perturbation limit, obtained from (Eqs. (7) and (C.17) of Ref. 7, are also plotted. From comparison with the GL results [12] we find that the MLF energy almost coincides with the exact energy in the entire regime of \( g \). The weak-coupling method fails for large \( g \) whereas the strong-coupling perturbation expansion of Ref. 8 breaks down and the LF perturbation result deviates much from the exact energy for low \( g \) (< 1.4).

To find out the energy dispersion within the MLF method the energy \( E_k \) has been calculated up to the second-order perturbation. The second-order perturbation correction to \( E_k \) within the MLF approach is obtained as

\[
E_k^{(2)} = - \frac{2g^2}{\omega} \left[ \frac{(3 + r)}{(1 + r)} (\omega t_e - t_e (1 - r)^2 \cos(ka))^2 \right] + \frac{(1 - r)^3 t_e^2 \sin^2(ka)}{(1 + r)} \]

\[
= - \frac{1}{N} \sum_l \sum_{n_r \geq 2} t_l^2 \frac{2^{2n_l} (X^2 + Y^2 + 2XY \cos(2ka))}{(n_l \omega)} (\sin \omega t_e (1 - r)^2 \cos(ka))^2 \]

where \( X = (-1)^n L^A \) and \( Y = (-1)^n R^B \), \( n_L, n_R, \) and \( A \) and \( B \) are defined in Eq. (9).

The corresponding expression for \( E_k^{(2)} \) within the LF approach is

\[
E_k^{(2)} = - \frac{2g^2}{\omega} \left[ \sum_{n=1}^{\infty} \frac{g^{2n}}{n!} \frac{\cos^2(ka)}{t_e - e^{-\frac{\omega}{k}} \sum_{m=1}^{\infty} \frac{g^{2(n+m)}}{(n+m)!} \frac{\sin^2(ka)}{t_e - e^{-\frac{\omega}{k}}} \right]
\]

where \( t_e = t e^{-\frac{\omega}{k}} \).

We have calculated \( E_k^{(2)} \), hence \( E_k \) up to the second-order perturbation within the LF and MLF approach. The second-order correction \( E_k^{(2)} \) and the ratio \( E_k^{(2)}/E_k \) for different \( k \) values are listed in Table I for several values of \( g \) for both the MLF and the LF approach for \( t = \omega = 1 \).

The following features are evident from our study:

(i) The second-order MLF corrections are appreciably smaller than the corresponding LF corrections for low values of \( g \) and \( k \). (ii) For small and intermediate values of \( g \) (e.g., \( g = 0.5 \) and 1.0) the MLF correction increases with increasing \( k \), however the MLF corrections are less than the corresponding LF values in the entire region of \( k \) except at \( ka = \pi/2 \) where both the MLF and LF corrections are same. In fact, for \( ka = \pi/2 \) the value of \( r \) becomes zero within the present MLF approach so the MLF basis reduces to the LF basis for \( ka = \pi/2 \).

(iii) The second-order perturbation correction within the LF method is symmetric around \( ka = \pi/2 \) as evident from Eq. (13). It is also minimum for \( ka = \pi/2 \). (iv) For small and intermediate values of \( g \) the second-order LF corrections are quite high. As a result \( E_k \) up to the second-order correction within the LF may not be an accurate estimate of \( E_k \). Nevertheless we have cited the LF values for comparison with the MLF values. (v) In a range \( 1.9 < g < 2.2 \) (for \( t = \omega = 1 \)) the LF second-order corrections are slightly less than the corresponding MLF corrections. For higher values of \( g \) the LF phonon basis reduces to the LF phonon basis and the results by these two methods become equivalent. (vi) With increasing \( g \) the second-order LF correction decreases considerably and becomes more and more \( k \)-independent. \( E_k^{(2)}/E_k \) within LF is around 0.027 for \( g = 2.5 \), for the whole range of \( k \).

In Fig. 3a we plot \( E_k \) vs. \( ka/\pi \) for \( g = 0.5 \) and 1.0 for the MLF approach and in Fig. 3b a comparison with LF result and the numerically exact results [12] [13] [14] is shown for \( g = 1 \). The MLF result is close to the numerically exact results only for low \( k \) but deviates considerably from the exact curve for higher values of \( k \). In this region the second-order corrections are increasingly higher implying that higher-order corrections are to be included to obtain accurate results. The LF re-
result is much less satisfactory than the MLF result for $g = 1$. The MLF perturbation method predicts a flat band (FIG. 3(b)) in the region $0.7\pi \leq ka \leq \pi$ which is consistent qualitatively with the exact result [12, 14] for $g = 1.0$. However, the values of $E_k$ predicted by the MLF in this region is found to be not consistent with the exact results.

For large $g$ the LF second-order corrections are small in the entire region of $k$, hence the dispersion given by the LF up to the second-order should be satisfactory. However, no numerical result for $E_k$ for large $g$ is available to us for a comparison.

In summary, in this paper we have proposed a simple MLF phonon basis containing a single variational parameter but capable of describing lattice distortions at distant sites from the charge carrier. For low and intermediate values of $g$ the second-order perturbation correction to the energy within this approach is much smaller than that corresponding to the LF approach. It appears that the ZPA with this MLF basis gives much more accurate results than that of ZPA with the LF basis. One representative calculation of effective mass is presented which corroborates this feature. The ground-state ($k = 0$) energy, calculated up to the second-order within the MLF approach agrees well with exact results for the entire range of coupling strength. For $g \leq 1.0$, the second-order correction within the MLF is small for low $k$ values and in this region the values of $E_k$ (band dispersion) predicted by the MLF is consistent with the numerically exact result. For large $g$ the LF perturbation corrections are small and the band dispersion predicted by the LF method is expected to be satisfactory. Computation of higher-order corrections would shed light on this issue.

Electronic address for correspondence: moon@cmp.saha.ernet.in

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