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Theory of Molecular Deformation Due to the Vibronic Interaction between Two Closely-spaced Electronic Levels. II. Green's Function Treatment

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Vibronic interaction between two closely-spaced electronic levels was investigated by use of the temperature Green's function. A Green's function version for the mean field approximation was derived which is capable of reproducing results obtained in the preceding work. The spectrum of phonon in the normal phase was then calculated and the critical (strictly speaking, characteristic, since we are dealing with a finite system) temperature T_c was obtained where the phonon becomes completely soft causing lattice distortion. The fluctuation effect was investigated and the mean field theory was examined for its applicability. When the level spacing is comparable to or larger than the value of the electron-phonon coupling constant, the mean field theory is satisfactory, but when the two levels are nearly degenerate, it is not so good because the problem in question is in the regime involving extremely strong coupling.

In the preceding work (referred to as I hereinafter) we have developed the mean field theory for the title subject. The model considered there is outlined as follows. Let a_i^+ (a_i) be the creation (annihilation) operator of the electron at level i, and $b^+(b)$ the creation (annihilation) operator of phonon. To simplify formulations we introduce the notations

$$C^+ = (a_1{}^+ \; a_2{}^+), \; C = \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}, \tag{1}$$

$$\phi = b^+ + b. \tag{2}$$

Thus, the Hamiltonian is given by

$$H = -\varepsilon C^+\sigma_3 C + g\phi C^+\sigma_1 C + \omega b^+ b, \tag{3}$$

where

$$\sigma_0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \; \sigma_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \; \sigma_2 = \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix}, \; \sigma_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{4}$$

are used. Later we will use the following marix for electron-phonon (vibronic) interaction:

$$\hat{g}\hat{\varepsilon} = \begin{pmatrix} 0 & g \\ g & 0 \end{pmatrix} = g\sigma_1. \tag{5}$$

Within the mean field approximation, operators b^+ and b in Eq. 3 are replaced by their averages. This approximation reduces the Hamiltonian given by Eq. 3 to

$$H^{eff} = -\varepsilon C^+\sigma_3 C + g\langle\phi\rangle C^+\sigma_1 C + \omega|\langle b\rangle|^2. \tag{6}$$

This form is based on an expectation that the so-called coherent state impling $\langle\phi\rangle \neq 0$ will be arranged by the electron-phonon interaction. In I H^{eff} was diagonalized by a linear canonical transformation. The value of $\langle\phi\rangle$ has been determined on the condition that the free energy of our system is optimized with respect to $\langle\phi\rangle$. As a numerical example, the bond distortion and stabilization energy were calculated for cyclobutadiene, in good agreement with those obtained by a current theory.

In this paper, we will present the Green's function theory for the problem. In the first section, we will show that the result obtained in I is derivable on the basis of the simplest approximation on Green's function. In the remaining section, we will investigate the fluctuation effect to examine the validity of the mean field theory.

Phonon Softening

It is known well in the physics of one-dimensional (1-D) conductors[4,5] that the phonon softening induced by electron-phonon interaction causes lattice deformation in these systems. To show that an analogous mechanism is operative in the present problem, we will begin with investigating behaviors of the phonon propagator in the normal state; we will work with the simplest approximation for the polarization part of the phonon propagator as shown in Fig. 1. Analytically it is expressed by

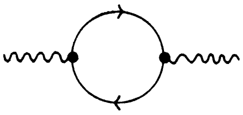

Fig. 1. First order polarization part of the phonon Green's function.

$$\Pi(\mathrm{i}\nu_n) = T\sum_{\mathrm{i}\omega_n} \mathrm{Tr}\, \hat{g}G^0(\mathrm{i}\omega_n)\hat{g}G^0(\mathrm{i}\nu_n - \mathrm{i}\omega_n), \tag{7}$$

where $G^0(\mathrm{i}\omega_n)$ is the Matsubara (temperature) Green's function[2] for a free electron and its expression can be obtained from the first term of Eq. 3 as

$$\begin{aligned} G^0(\mathrm{i}\omega_n) &= (\mathrm{i}\omega_n\sigma_0 + \varepsilon C^+\sigma_3 C)^{-1} \\ &= \begin{pmatrix} \dfrac{1}{\mathrm{i}\omega_n + \varepsilon} & 0 \\ 0 & \dfrac{1}{\mathrm{i}\omega_n - \varepsilon} \end{pmatrix} \\ &= \sum_k G_k^0(\mathrm{i}\omega_n)\,\sigma_k \end{aligned} \tag{8}$$

with

$$\begin{aligned} G_0^0(\mathrm{i}\omega_n) &= \frac{1}{2}\left(\frac{1}{\mathrm{i}\omega_n + \varepsilon}\right) + \left(\frac{1}{\mathrm{i}\omega_n - \varepsilon}\right) \\ G_3^0(\mathrm{i}\omega_n) &= \frac{1}{2}\left(\frac{1}{\mathrm{i}\omega_n + \varepsilon} - \frac{1}{\mathrm{i}\omega_n - \varepsilon}\right). \end{aligned} \tag{9}$$

If we use Eqs. 5 and 8, Eq. 7 is rewritten as

$$\Pi(\mathrm{i}\nu_n) = 2g^2\, T\sum_{\mathrm{i}\omega n} \{G_0^0(\mathrm{i}\omega_n)\, G_0^0(\mathrm{i}\nu_n - \mathrm{i}\omega_n) - G_3^0(\mathrm{i}\omega_n)G_3^0(\mathrm{i}\nu_n - \mathrm{i}\omega_n)\}. \tag{10}$$

The frequency sum[3] in Eq. 10 leads to

$$\Pi\,(\mathrm{i}\nu_n) = -\, g^2 \frac{4\varepsilon}{\nu_n^2+4\varepsilon^2} \tanh(\varepsilon/2T). \tag{11}$$

The interacting phonon Green's function is obtained from the Dyson equation

$$D(\mathrm{i}\nu_n) = D^0(\mathrm{i}\nu_n)/\{1 - D^0(\mathrm{i}\nu_n)\Pi(\mathrm{i}\nu_n)\}, \tag{12}$$

where the free phonon propagator $D^0(\mathrm{i}\nu_n)$ is given by

$$D^0(\mathrm{i}\nu_n) = \frac{-2\omega}{\nu_n^2+\omega^2}. \tag{13}$$

Combining Eqs. 11, 13, and 12, we obtain

$$D(\mathrm{i}\nu_n) = \frac{-2\omega(\nu_n^2+4\varepsilon^2)}{(\nu_n^2+\omega^2)(\nu_n^2+4\varepsilon^2)-8g^2\,\varepsilon\omega\tanh(\varepsilon/2T)}. \tag{14}$$

In this equation $\mathrm{i}\nu_n$ is analytically continued to ν and thus the poles of Eq. 14 are found at

$$(\nu^2)_\pm = \frac{1}{2}(\omega^2 + 4\varepsilon^2) \pm \left\{\left(\frac{\omega^2+4\varepsilon^2}{2}\right)^2 + 8g^2\varepsilon\omega\tanh(\varepsilon/2T) - 4\varepsilon^2\,\omega^2\right\}^{1/2}. \tag{15}$$

First, we note that, if coupling constant g is extremely small, Eq. 15 reduces to

$$(\nu^2)_\pm \simeq \begin{cases}\omega^2 \\ 4\varepsilon^2\end{cases} \text{for } \omega>\varepsilon, \tag{16a}$$

and

$$(\nu^2)_\pm \simeq \begin{cases}4\varepsilon^2 \\ \omega^2\end{cases} \text{for } \omega<\varepsilon. \tag{16b}$$

They are spectra of independent electron and phonon. Aside from this simplest case, spectrum ν_- in general depends on temperature in a very specific way. In paticular, the behavior of ν_- is quite specific. If g is strong enough, it becomes soft as temperature is lowered. Complete softening of ν_- occurs at the temperature T_c in accordance with

$$\tanh(\varepsilon/2T_c) = (\varepsilon\omega/2g^2). \tag{17}$$

A unique solution is obtainable for T_c if g is so strong as to satisfy the inequality

$$g^2 > \varepsilon\omega/2. \tag{17a}$$

At the temperature T_c we get the spectrum

$$(\nu^2)_\pm = \begin{cases}\omega^2+4\varepsilon^2 \\ 0\end{cases} \tag{18}$$

Equation 17 is equal to the relation defining the characteristic temperature for the distortive transition (given by Eq. 38 in I).

The above-demonstrated similarity to the case of 1-D conductor is of significance; however, it is worth while to take notice of the strong coupling condition (17a) applicable to our case, in contrast to the case of 1-D Peierls transition where any weak coupling can lead to the softening.

Electron Propagator

The description in the preceding section is inapplicable to the temperature range below the characteristic temperature T_c. To see how the system behaves at $T \lesssim T_c$, we consider the solution with deformation on the basis of the effective Hamiltonian (6). The propagator for a "free" electron suitable for this case is given by

$$G(\mathrm{i}\omega_n) = (\mathrm{i}\omega_n\sigma_0 + \varepsilon\sigma_3 - \Delta\sigma_1)^{-1}, \tag{19}$$

where

$$\Delta = g\langle\phi\rangle. \tag{20}$$

A straightforward calculation leads to an alternative form for Eq. 19

$$G(\mathrm{i}\omega_n) = \sum_k G_k(\mathrm{i}\omega_n)\sigma_k \tag{21}$$

$$G_k(\mathrm{i}\omega_n) = \frac{1}{(\mathrm{i}\omega_n-\tilde{\varepsilon})(\mathrm{i}\omega_n+\tilde{\varepsilon})} \times \begin{cases}\mathrm{i}\omega_n \\ \Delta \\ 0 \\ -\varepsilon\end{cases} \text{for } k = \begin{cases}0 \\ 1 \\ 2 \\ 3\end{cases}, \tag{21a}$$

where

$$\tilde{\varepsilon} = \sqrt{\varepsilon^2+\Delta^2}. \tag{22}$$

To estimate $\langle\phi\rangle$, we now look for the solution of the static distortion as has been done in I, namely,

$$-\left\langle\frac{\partial}{\partial\tau} b\right\rangle = \langle[b,H]\rangle = 0, \tag{23}$$

which is the average of the Heisenberg equation of motion with respect to the imaginary time τ. Using Eq. 3 to make a calculation on Eq. 23, we obtain

$$\langle b\rangle = -\frac{g}{\omega}\langle C^+\sigma_1 C\rangle. \tag{24}$$

Obviously, for $\langle b^+\rangle$ we are led to entirely the same expression and it follows that

$$\Delta = g\langle\phi\rangle = -\frac{2g^2}{\omega}\langle C^+\sigma_1 C\rangle = \frac{g^2}{\omega}G_1(0^-), \tag{25}$$

where 0^- is negative infinitesimal and

$$\begin{aligned} G_1(0^-) &= T\sum_{\mathrm{i}\omega n} G_1(\mathrm{i}\omega_n)\exp\,(\mathrm{i}\omega_n 0^-) \\ &= T\sum_{\mathrm{i}\omega n} \frac{\Delta}{(\mathrm{i}\omega_n-\tilde{\varepsilon})(\mathrm{i}\omega_n+\tilde{\varepsilon})}\exp\,(\mathrm{i}\omega_n\, 0^-) \\ &= (\Delta/2\tilde{\varepsilon})\tanh\,(\tilde{\varepsilon}/2T). \end{aligned} \tag{26}$$

Combining Eqs. 25 and 26, we arrive at the "gap" equation

$$(\tilde{\varepsilon}\omega/2g^2) = \tanh\,(\tilde{\varepsilon}/2T), \tag{27}$$

which is identical with Eq. 36 in I.

Thermodynamics

According to Ref. 6 the difference in free energy between the coherent and normal phases

$$\delta F = F - F_n \tag{28}$$

is given by

$$\delta F = \int_0^g \mathrm{d}g\,\langle\phi C^+\,\sigma_1 C\rangle. \tag{29}$$

In the integrand in Eq. 29 the average $\langle\phi C^+\sigma_1 C\rangle$ is approximated to $\langle\phi\rangle\ \langle C^+\sigma_1 C\rangle$, which permits Eq. 29 with the aid of Eqs. 25, 26, and 27 to give

$$\begin{aligned} \delta F &= -\int_0^g \mathrm{d}g\, 2\langle\phi\rangle\frac{\Delta}{2\tilde{\varepsilon}}\tanh\,(\tilde{\varepsilon}/2T) \\ &= -\int_0^g \mathrm{d}g\,(\omega\Delta^2/2g^3). \end{aligned} \tag{29a}$$

The integration for Eq. 29a can be performed as follows:

$$\begin{aligned}\delta F &= \int_0^g \frac{\omega\Delta^2}{2}\,\mathrm{d}\left(\frac{1}{2g^2}\right)\\ &= \frac{\omega}{2}\int_\varepsilon^{\tilde\varepsilon} \Delta^2 \frac{\mathrm{d}}{\mathrm{d}\tilde\varepsilon}\left(\frac{1}{2g^2}\right)\mathrm{d}\tilde\varepsilon\\ &= \frac{\omega}{2}\int_\varepsilon^{\tilde\varepsilon} \Delta^2 \frac{\mathrm{d}}{\mathrm{d}\tilde\varepsilon}\left[\frac{1}{\tilde\varepsilon\omega}\tanh(\tilde\varepsilon/2T)\right]\mathrm{d}\tilde\varepsilon\\ &= \frac{1}{2}\int_\varepsilon^{\tilde\varepsilon} (\tilde\varepsilon^2-\varepsilon^2)\frac{\mathrm{d}}{\mathrm{d}\tilde\varepsilon}\left[\frac{1}{\tilde\varepsilon}\tanh(\tilde\varepsilon/2T)\right]\mathrm{d}\tilde\varepsilon, \qquad (30)\end{aligned}$$

where for the second line the gap equation (27) has been used and for the third line the definition of $\tilde\varepsilon$ by Eq. 22 used. Finally, the integration by parts for Eq. 30 gives the free energy difference as

$$\begin{aligned}\delta F &= \frac{1}{2}\left[\frac{\tilde\varepsilon^2-\varepsilon^2}{\tilde\varepsilon}\tanh(\tilde\varepsilon/2T)\right]\\ &\quad -2T\{\ln[2\cosh(\tilde\varepsilon/2T)] - \ln[2\cosh(\varepsilon/2T)]\}. \qquad (31)\end{aligned}$$

For comparison we evaluate the free energy for the normal state; the partition function for this state is given by

$$\begin{aligned}Z_n &= \mathrm{Tr}\exp(-\varepsilon C^+\sigma_1 C/T)\\ &= \mathrm{Tr}\exp(-\varepsilon a_1^+a_1/T)\times\mathrm{Tr}\exp(\varepsilon a_2^+a_2/T)\\ &= [\exp(\varepsilon/2T)+\exp(-\varepsilon/2T)]^2 \qquad (32)\end{aligned}$$

for which we have made use of the fact that the eigenvalue of $a_i^+a_i$ is 0 or 1. Therefore, the free energy of the normal state is given by

$$F_n = -T\ln Z_n = -2T\ln[2\cosh(\varepsilon/2T)], \qquad (33)$$

which, combined with Eq. 31, leads to

$$F = (\Delta^2\omega/4g^2) - 2T\ln[2\cosh(\tilde\varepsilon/2T)]. \qquad (34)$$

Thus, we have obtained the free energy of the distorted state, which is identical with Eq. 33 in I; as far as the above treatment is concerted, the two methods have been shown to be identical.

Fluctuation Effect

Let us examine to what extent the above results are valid by estimating the correction due to fluctuation. To describe the deviation from the mean field quantities, new operator β, β^+, and ϕ are defined by

$$b = \langle b\rangle + \beta,\; b^+ = \langle b\rangle + \beta^+, \qquad (35)$$

$$\phi = \beta + \beta^+. \qquad (36)$$

These definitions allow the Hamiltonian to be rewritten as

$$H = H_0 + H_{\mathrm{int}} + \omega\langle b\rangle\phi + \omega\beta^+\beta + \omega|\langle b\rangle|^2, \qquad (37)$$

with

$$H_0 = C^+(-\varepsilon\sigma_3 + \Delta\sigma_1)C, \qquad (38)$$

$$H_{\mathrm{int}} = C^+ g\phi\sigma_1 C. \qquad (39)$$

In what follows, we attempt to estimate the fluctuation effect on electronic spectra within the lowest order of g by considering the self-energy contribution shown in Fig. 2.

Mean field electron propagator G (displayed in solid line in Fig. 2) is obtained from H_0 in Eq. 37. It has already been given by Eq. 21 with Δ to be determined by Eq. 27. Phonon propagator D^0 (displayed in wavy line in Fig. 2) is easily calculated from the third and fourth terms of Eq. 37 and is shown to be identical with Eq. 13.

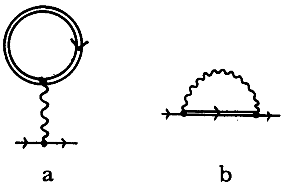

Fig. 2. First order self-energy terms of the electron propagator.
a: Direct term, b: exchange term.

Using these expressions for propagators, we can express the self-energy parts in Fig. 2 analytically as

$$\Sigma = \Sigma_H + \Sigma_E, \qquad (40)$$

where

$$\Sigma_H(\mathrm{i}\omega_n) = Tg^2\sigma_1 D^0(0)\sum_{\mathrm{i}\nu_n}\mathrm{Tr}\,\sigma_1 G(\mathrm{i}\nu_n) \qquad (40\mathrm{a})$$

$$\Sigma_E(\mathrm{i}\omega_n) = -Tg^2\sum_{\mathrm{i}\nu_n}\sigma_1 G(\mathrm{i}\nu_n)D^0(\mathrm{i}\omega_n - \mathrm{i}\nu_n)\sigma_1. \qquad (40\mathrm{b})$$

Σ_H is the contribution of Fig. 2a and Σ_E that of Fig. 2b. Following the usual procedure, we evaluate Σ_H as

$$\begin{aligned}\Sigma_H(\mathrm{i}\omega_n) &= (-4g^2/\omega)T\sum_{\mathrm{i}\nu_n}\sigma_1 G_1(\mathrm{i}\nu_n)\\ &= \sigma_1(4g^2/\omega)(\Delta/2\tilde\varepsilon)\tanh(\tilde\varepsilon/2T)\\ &= \sigma_1\Delta. \qquad (41)\end{aligned}$$

In the last step in Eq. 41 we have used the gap equation (27).

The calculation of Σ_E is tedious: First, the algebra of the Pauli matrices yields

$$\Sigma_E = \sigma_0\Sigma_0 + \sigma_1\Sigma_1 - \sigma_3\Sigma_3, \qquad (42)$$

where

$$\Sigma_k(\mathrm{i}\omega_n) = -g^2T\sum_{\mathrm{i}\nu_n}G_k(\mathrm{i}\nu_n)D^0(\mathrm{i}\omega_n - \mathrm{i}\nu_n),$$

$$k = 0, 1, 3. \qquad (42\mathrm{a})$$

Each term of this equation will be evaluated separately. The frequency summation of Σ_0 is performed to give

$$\Sigma_0(\mathrm{i}\omega_n) = g^2\left[\frac{\mathrm{i}\omega_n N_1}{(\mathrm{i}\omega_n)^2-(\tilde\varepsilon+\omega)^2} + \frac{\mathrm{i}\omega_n N_0}{(\mathrm{i}\omega_n)^2-(\tilde\varepsilon-\omega)^2}\right], \qquad (43)$$

where

$$\begin{aligned}N_1 &= n(-\tilde\varepsilon) + n_b(\omega)\\ N_0 &= 1 - n(-\tilde\varepsilon) + n_b(\omega) \qquad (44)\end{aligned}$$

and $n(-\tilde\varepsilon)$ and $n_b(\omega)$ are, respectively, the Fermi and Bose function with corresponding arguments.

We have an approximation

$$(\mathrm{i}\omega_n)^2 - (\tilde\varepsilon+\omega)^2 \simeq \tilde\varepsilon^2 - (\tilde\varepsilon+\omega)^2 \simeq -2\tilde\varepsilon\omega \qquad (45)$$

considering that $\tilde\varepsilon \gg \omega$; this approximation is possibly applicable to the example case discussed in I. Equation 43 can then be reduced to

$$\Sigma_0(\mathrm{i}\omega_n) \simeq \Sigma_0(\pm\tilde\varepsilon) = \mp\tilde\varepsilon/4. \qquad (46)$$

The remaining two terms in Eq. 42 are evaluated in a similar manner as

$$\begin{aligned}\Sigma_1(\mathrm{i}\omega_n) &= -\Delta/4\\ \Sigma_3(\mathrm{i}\omega_n) &= \varepsilon/4. \qquad (47)\end{aligned}$$

Combining Eqs. 41, 46, and 47, we obtain

$$\Sigma(\mathrm{i}\omega_n) \simeq \Sigma(\pm\tilde\varepsilon) = \frac{1}{4}\begin{pmatrix}\mp\tilde\varepsilon-\varepsilon & 3\Delta\\ 3\Delta & \mp\tilde\varepsilon+\varepsilon\end{pmatrix}. \qquad (48)$$

Finally, the full electron propagator

$$G(\mathrm{i}\omega_n) = [G_0^{-1}(\mathrm{i}\omega_n) - \textstyle\sum(\mathrm{i}\omega_n)]^{-1}$$

is given in the form

$$G(\mathrm{i}\omega_n) \simeq \begin{pmatrix} \mathrm{i}\omega_n \pm \frac{\bar{\varepsilon}}{4} + \frac{5\varepsilon}{4} & -\frac{7\Delta}{4} \\ -\frac{7\Delta}{4} & \mathrm{i}\omega_n \pm \frac{\bar{\varepsilon}}{4} - \frac{5\varepsilon}{4} \end{pmatrix}^{-1}. \tag{49}$$

The electronic spectrum which involves the contribution from the fluctuation effect is determined from Eq. 49 as

$$\nu = \pm\,\bar{\varepsilon}\left(1 + \frac{24}{25}\frac{\Delta^2}{\bar{\varepsilon}^2}\right)^{1/2}. \tag{50}$$

According to this equation the electronic spectrum is to be corrected by a factor

$$\lambda = \left(1 + \frac{24}{25}\frac{\Delta^2}{\bar{\varepsilon}^2}\right)^{1/2} - 1. \tag{51}$$

In the case where the two electronic levels are nearly degenerate, *i.e.*, $\varepsilon \simeq 0$, the factor (51) is considerably large since $\lambda \lesssim \sqrt{2} - 1$, whereas if ε is so large that $\bar{\varepsilon} \gg \Delta$ the correction is small since $\lambda \simeq 0$. The latter inequality implies

$$\varepsilon \simeq 2g^2/\omega, \tag{52}$$

i.e., relatively weak coupling cases. Such cases as involve nearly degenerate levels belong to the extremely strong coupling regime.

Conclusion

We have made use of Green's functions to reproduce the results from the mean field theory presented in I and examined effects of fluctuation on electronic spectra. We are led to conclude that, as seen from Eq. 50, the mean field theory is not necessarily good; if the level spacing is fairly large, *i.e.*, $\varepsilon \simeq g$ or $\varepsilon \simeq \Delta$, the mean field theory is a pretty good approximation, but if the two levels are nearly degenerate, a substantial deviation is inevitable to the mean field theory. For this reason, the numerical estimate for cyclobutadiene (where $\varepsilon \simeq 0$) given in I, though in good agreement with results of other works, must be considered to be only qualitatively reliable.

In order to improve this situation and to reach better understanding of this problem, the perturbation scheme can be hardly suitable because we have no smaller parameters available for the series expansion. In paticular, the vertex renormalization which we have ignored can be expected to be superior to any other corrections. For metals we have the well-known Migdal theorem⁷⁾ which has demonstrated that the vertex correction is as small as the order of the small parameter m/M (electron-nucleus mass ratio). We do not think that the theorem is actually applicable to the case of finite systems such as molecules.

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