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STRONG COUPLING DIAGRAMMATIC APPROACH TO THE ANDERSON-HOLSTEIN HAMILTONIAN

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We develop a strong-coupling approach to investigate the Anderson-Holstein model with strong repulsion on impurity centers. We derive the relation between electron propagators and correlation functions and prove that for the impurity electrons the latter is identical to the mass operator of the conduction electrons. Strong electron-phonon interaction determines formation of polarons with heavy clouds of phonons surrounding impurity electrons. We demonstrate the existence of a collective excitation mode of these clouds and obtain the dependence of its energy on the hybridization of the impurity with band states. Hybridization is shown to cause softening of the collective mode and its total suppression at sufficiently large values of the hybridization parameter.

Key words: strongly correlated electron system, Dyson equation, Green's function, polaron, phonon clouds.

1. INTRODUCTION

Strong correlation effects can occur in metallic systems due to both strong electron-electron interactions and strong electron-phonon coupling, including their interplay as well [1].

In many-body systems strongly correlated electrons are also strongly coupled to the lattice vibrations, for example in V_2O_3 [2, 3], manganites [4], and fullerides [5]. The strong electron-electron interactions can be described by the Hubbard [6] and Anderson [7] models. The Holstein model [8] has been used to examine electron-phonon interactions. The Anderson-Holstein model includes both types of interaction.

The advances in the field of molecular electronics have revived the interest to the problem of electron-phonon interaction because electron-vibrational coupling within the molecule is important for understanding the properties of such devices. From the theoretical point of view it is the problem of small polaron discussed by Holstein [8].

The Hamiltonian of the Anderson-Holstein impurity model has the following form

$$H = H_{el} + H_{ph} + H_{hyb} + H_{el\text{-}ph},\ H_{el} = \sum_{\mathbf{k}\sigma} \varepsilon(\mathbf{k}) C^{+}_{\mathbf{k}\sigma} C_{\mathbf{k}\sigma} + \sum_{\sigma} \varepsilon^{0}_{f}(\mathbf{k}) f^{+}_{\sigma} f_{\sigma} + U_0 n_{\uparrow} n_{\downarrow},$$
$$H_{ph} = h\omega_0 (a^{+}a + \frac{1}{2}),\ q = \frac{1}{\sqrt{2}}(a + a^{+}),\ p = \frac{i}{\sqrt{2}}(a^{+} - a), \tag{1}$$
$$H_{hyb} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}\sigma} (V_{\mathbf{k}} f^{+}_{\sigma} C_{\mathbf{k}\sigma} + V^{*}_{\mathbf{k}} C^{+}_{\mathbf{k}\sigma} f_{\sigma}),\ H_{el-ph} = \bar{g} q n,\ n = \sum_{\sigma} n_{\sigma},\ n_{\sigma} = f^{+}_{\sigma} f_{\sigma},$$

where q and p are local coordinate and momentum of the optical phonons, ω_0 their frequency, $a(a^{+})$, $C_{\mathbf{k}\sigma}(C^{+}_{\mathbf{k}\sigma})$, and $f_{\sigma}(f^{+}_{\sigma})$ annihilation (creation) operators of phonons, conduction, and impurity electrons, correspondingly. $\varepsilon(\mathbf{k})$ is the kinetic energy of conduction electrons and ε^{0}_{f} is the local impurity energy, which are calculated from the chemical potential μ_0 of the system: $\varepsilon(\mathbf{k}) = \xi(\mathbf{k}) - \mu_0$, $\varepsilon^{0}_{f} = \bar{\varepsilon}^{0}_{f} - \mu_0$. U_0 is the on site Coulomb repulsion of impurity electrons with opposite spins, $V_{\mathbf{k}}$ is the

matrix element of hybridization, N is the number of lattice sites, and $\bar{g}$ is the constant of electron-phonon interaction. Both quantities, U_0 and $\bar{g}$, are far too large to be treated by perturbation theory and should be included in the zero-th order Hamiltonian $H_0 = H_{el} + H_{ph}$.

To eliminate the linear q-term of Hamiltonian we use the Lang-Firsov [9] transformation

$$\tilde{H} = \mathrm{e}^{S} H \mathrm{e}^{-S},\ S = -\mathrm{i}\, g n p,\ g = \frac{\bar{g}}{\hbar\omega_0}. \tag{2}$$

The transformed impurity operators become: $\tilde{f}_\sigma = \mathrm{e}^{S} f_\sigma \mathrm{e}^{-S} = \mathrm{e}^{\mathrm{i}gp} f_\sigma$, $\tilde{f}_\sigma^{+} = f_\sigma^{+} \mathrm{e}^{-\mathrm{i}gp}$, while conduction electron operators remain unchanged. As a result we obtain $\tilde{H} = H_0 + H_{\mathrm{int}}$, where

$$H_0 = \sum_{\mathbf{k}\sigma} \varepsilon(\mathbf{k}) C_{\mathbf{k}\sigma}^{+} C_{\mathbf{k}\sigma} + \sum_{\sigma} \varepsilon_f n_\sigma + U n_\uparrow n_\downarrow + \hbar\omega_0 (a^{+}a + \frac{1}{2}) \tag{3}$$

and

$$H_{\mathrm{int}} = \sum_{\sigma} (\tilde{f}_\sigma^{+} b_\sigma + b_\sigma^{+} \tilde{f}_\sigma). \tag{4}$$

Here

$$\begin{gathered} \varepsilon_f = \varepsilon_f^0 - \mu,\ \mu = \mu_0 + \alpha\hbar\omega_0,\ U = U_0 - 2\alpha\hbar\omega_0,\ \alpha = \frac{g^2}{2}, \\ b_\sigma = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} C_{\mathbf{k}\sigma} V_{\mathbf{k}},\ b_\sigma^{+} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} V_{\mathbf{k}}^{*} C_{\mathbf{k}\sigma}^{+}. \end{gathered} \tag{5}$$

The operator $b_\sigma (b_\sigma^{+})$ represents a local conduction electron and $\tilde{f}_\sigma (\tilde{f}_\sigma^{+})$ stands for an impurity electron, surrounded by phonons, i.e., a *polaron*.

In the perturbation approach developed below we shall use the *generalized Wick theorem* proposed in [10-15] for strongly correlated systems. The generalized theorem will be employed for the impurity subsystem and the standard theorem will be used for conduction electrons and optical phonons.

In section 2 we introduce the finite temperature Matsubara Green's functions for the conduction and impurity electrons in the interaction representation. In Section 3 we analyze the dynamics of phonon clouds. In section 4 we provide explicit examples of diagram calculation for the full propagators and section 5 contains the discussion of the results and conclusions.

2. DIAGRAMMATIC APPROACH

The Matsubara renormalized Green's functions of conduction and impurity electrons in interaction representation are defined as follows:

$$\begin{aligned} &G(\sigma\tau \mid \sigma'\tau') = -\left\langle T b_\sigma(\tau) \bar{b}_{\sigma'}(\tau') U(\beta) \right\rangle_0^c,\ \ g_e(\sigma\tau \mid \sigma'\tau') = -\left\langle T f_\sigma(\tau) \bar{f}_{\sigma'}(\tau') U(\beta) \right\rangle_0^c, \\ &g_p(\sigma\tau \mid \sigma'\tau') = -\left\langle T \tilde{f}_\sigma(\tau) \tilde{\bar{f}}_{\sigma'}(\tau') U(\beta) \right\rangle_0^c, \end{aligned} \tag{6}$$

where $\tau,\ \tau'$ stand for the imaginary time with $0 < \tau < \beta$, were β is the inverse temperature of the system. Here T is the time ordering operator. Statistical averaging $\langle \ldots \rangle_0^c$ is done with the zero-th order density operator of the grand canonical ensemble $\frac{\mathrm{e}^{-\beta H^0}}{\mathrm{Tr}\, \mathrm{e}^{-\beta H^0}}$. The superscript c in (6) indicates that only connected diagrams are taken into account; g_{e} and g_{p} are electron and polaron propagators. The evolution operator $U(\beta)$ is defined as usual: $U(\beta) = T \exp(-\int_0^\beta H_{\mathrm{int}}(\tau)\, \mathrm{d}\tau)$.

Besides of the normal Green's functions (6) we will also consider the anomalous Green's functions describing the superconducting pairing correlations

$$F(\sigma\tau|-\sigma'\tau') = -\left\langle Tb_{\sigma}(\tau)b_{-\sigma'}(\tau')U(\beta)\right\rangle_0^c, f_p(\sigma\tau|-\sigma'\tau') = -\left\langle T\tilde{f}_{\sigma}(\tau)\tilde{f}_{-\sigma'}(\tau')U(\beta)\right\rangle_0^c. \quad (7)$$

In addition, there exist also propagators of the phonon clouds (see paper [14])

$$\begin{aligned}&\Phi(\tau_1-\tau_2) = \left\langle T\exp[\mathrm{i}g(\mathrm{p}(\tau_1)-\mathrm{p}(\tau_2))]U(\beta)\right\rangle_0^c,\ \varphi(\tau_1-\tau_2) = \left\langle T\exp[\mathrm{i}g(\mathrm{p}(\tau_1)+\mathrm{p}(\tau_2))]U(\beta)\right\rangle_0^c,\\ &\Phi(\tau_1\tau_2|\tau_3\tau_4) = \left\langle T\exp[\mathrm{i}g(\mathrm{p}(\tau_1)+\mathrm{p}(\tau_2)-\mathrm{p}(\tau_3)-\mathrm{p}(\tau_4))]U(\beta)\right\rangle_0^c.\end{aligned} \quad (8)$$

We henceforth assume that the system is in the paramagnetic state. The Fourier representation of the phonon cloud propagator can then be obtained using Laplace approximation. In the *strong-coupling limit*, $\alpha \gg 1$, the propagator takes the following form [15,16]

$$\Phi(\mathrm{i}\Omega) = \frac{2\omega_c}{\Omega^2+\omega_c^2},\ \omega_c = \alpha\omega_0. \quad (9)$$

This expression describes the harmonic propagator of the collective mode of phonons belonging to the polaron cloud and having the collective frequency ω_c. Thus, Eq. (9) defines the concept of free collective oscillations of the phonon clouds surrounding the polarons.

The Laplace approximation for the strong-coupling limit $\alpha \gg 1$ also serves to prove the relation:

$$\Phi_0(\tau_1\tau_2|\tau_3\tau_4) \approx \Phi_0(\tau_1|\tau_3)\bar{\Phi}_0(\tau_2|\tau_4) + \bar{\Phi}_0(\tau_1|\tau_4)\Phi_0(\tau_2|\tau_3). \quad (10)$$

This equation and its generalization for the many time arguments are considered as the *Wick theorem* for phonon clouds in the strong coupling polaron regime.

3. THE DYNAMICS OF PHONON CLOUDS

We will now consider the renormalization of the phonon cloud propagators by the hybridization of impurity and band electron states. In the second and fourth order of perturbation theory we obtain the contributions shown in Fig. 1.

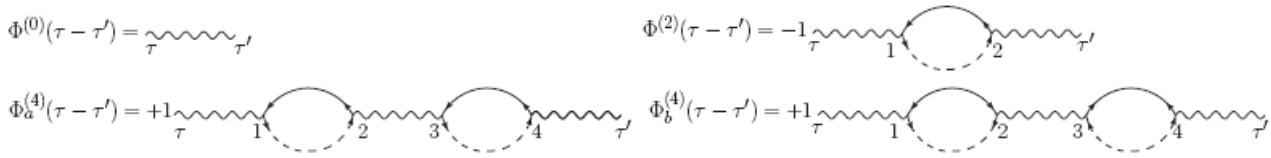

Fig. 1 – Renormalization of the phonon clouds contributions. Thin solid lines correspond to conduction electron propagators and dashed lines correspond to impurity electron propagators. The diagrams represent the simplest contributions of the second and fourth order of perturbation theory. The loops correspond to the simplest polarization operators.

The following expressions represent the contribution of some of the diagrams shown in the above figure:

$$\begin{aligned}&\Phi^{(2)}(\tau-\tau') = \iint \mathrm{d}\tau_1\mathrm{d}\tau_2\Phi^{(0)}(\tau-\tau_1)\Pi_2(\tau_1-\tau_2)\Phi^{(0)}(\tau_2-\tau'),\\ &\varphi^{(2)}(\tau-\tau') = \iint \mathrm{d}\tau_1\mathrm{d}\tau_2\Phi^{(0)}(\tau-\tau_1)\Pi_2^{\mathrm{s}}(\tau_1-\tau_2)\Phi^{(0)}(\tau'-\tau_2),\\ &\Phi^{(4)}(\tau-\tau') = \int\int\int\int \mathrm{d}\tau_1\mathrm{d}\tau_2\mathrm{d}\tau_3\mathrm{d}\tau_4\Phi^{(0)}(\tau-\tau_1)\Pi_2(\tau_1-\tau_2)\Phi^{(0)}(\tau_2-\tau_3)\Pi_2(\tau_3-\tau_4)\Phi^{(0)}(\tau_4-\tau'),\end{aligned} \quad (11)$$

where

$$\Pi_2(1-2) = -\left\langle Tb_1\bar{b}_2\right\rangle_0\left\langle Tf_2\bar{f}_1\right\rangle_0, \Pi_2^{\mathrm{s}}(1-2) = \left\langle Tb_1b_2\right\rangle_0\left\langle T\bar{f}_2\bar{f}_1\right\rangle_0, \bar{\Pi}_2^{\mathrm{s}}(1-2) = \left\langle T\bar{b}_1\bar{b}_2\right\rangle_0\left\langle Tf_2f_1\right\rangle_0. \quad (12)$$

In the normal state of the system all the anomalous Green's functions are equal to zero and we set to zero also the vanishing propagators φ and $\bar{\varphi}$.

The diagram series for the normal propagator $\Phi(\tau-\tau')$ can be put in the form of Dyson equation

$$\Phi(\tau-\tau')=\Phi^{(0)}(\tau-\tau')+\iint d\tau_1 d\tau_2 \Phi^{(0)}(\tau-\tau_1)\Pi(\tau_1-\tau_2)\Phi^{(0)}(\tau_2-\tau'), \tag{13}$$

where $\Pi(\tau)$ is the full polarization operator.

$$\Phi(i\Omega)=\Phi^{(0)}(i\Omega)+\Phi^{(0)}(i\Omega)\Pi(i\Omega)\Phi(i\Omega), \Phi(i\Omega)=\frac{\Phi^{(0)}(i\Omega)}{1-\Phi^{(0)}(i\Omega)\Pi(i\Omega)}=\frac{1}{(\Phi^{(0)}(i\Omega))^{-1}-\Pi(i\Omega)}. \tag{14}$$

In the Fourier representation we have
Using the expression (9) we then find

$$\Phi(i\Omega)=\frac{2\omega_c}{\Omega^2+\omega_c^2-2\omega_c\Pi(i\Omega)}. \tag{15}$$

The pole of this equation determines the renormalization of the collective phonon frequency ω_c:

$$E^2-\omega_c^2+2\omega_c\Pi(E)=0. \tag{16}$$

The simplest electron Green's functions have the form $(\overline{\sigma}=-\sigma)$:

$$\begin{gathered} G_\sigma^0(i\omega)=\frac{1}{N}\sum_k\frac{|V_k^2|}{i\omega-\varepsilon(k)}, \quad g_\sigma^0(i\omega)=\frac{1-n_{-\sigma}}{\lambda_\sigma(i\omega)}+\frac{n_{-\sigma}}{\overline{\lambda}_{\overline{\sigma}}(i\omega)}, \quad \lambda_\sigma(i\omega)=i\omega+E_0-E_\sigma, \\ \lambda_\sigma(i\omega)=i\omega+E_0-E_\sigma, \quad E_0=0,\ E_\sigma=\varepsilon_f, \quad E_2=U+2\varepsilon_f, \quad \overline{\lambda}_{\overline{\sigma}}(i\omega)=i\omega+E_{-\sigma}-E_2, \\ Z_0=e^{-\beta E_0}+2e^{-\beta E_{\overline{\sigma}}}+e^{-\beta E_2}, \quad n_{\overline{\sigma}}=\frac{e^{-\beta E_{\overline{\sigma}}}+e^{-\beta E_2}}{Z_0}. \end{gathered} \tag{17}$$

These functions allow to find the lowest order polarization operator of the normal state: $\Pi_2(i\Omega)=-\frac{1}{\beta}\sum_\omega G_\sigma^0(i\omega)g_\sigma^0(i\omega-i\Omega)$. Carrying out the summation over frequencies results in:

$$\begin{gathered} \Pi_2(i\Omega)=\frac{1}{N}\sum_{k\sigma}|V_k|^2\left\{\frac{1-n_{\overline{\sigma}}}{i\Omega-\varepsilon(k)+\varepsilon_f}\left[\frac{1}{e^{\beta\varepsilon(k)}+1}-\frac{1}{e^{\beta\varepsilon_f}+1}\right]+\right. \\ \left.+\frac{n_{\overline{\sigma}}}{i\Omega-\varepsilon(k)+U+\varepsilon_f}\left[\frac{1}{e^{\beta\varepsilon(k)}+1}-\frac{1}{e^{\beta(U+\varepsilon_f)}+1}\right]\right\}, \quad \frac{1}{N}\sum_k\ldots\to\int\ldots\rho(E)dE, \end{gathered} \tag{18}$$

where $\rho(E)$ is the density of states. From the equation (16), for the case $T=0$ and assuming a flat density of states $\rho(E)=\frac{1}{2W}\begin{cases}1, & E<|W|\\ 0, & E>|W|\end{cases}$, we can determine the dependence of the collective mode energy E on the hybridization parameter V (W is the half bandwidth). The results of numerical calculations are presented in Fig. 2.

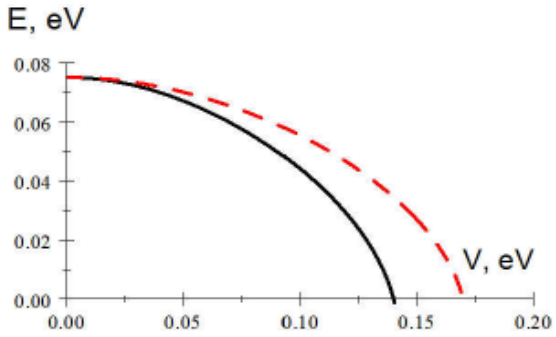

Fig. 2 – Energy of the collective mode vs hybridization V, for different band widths W = 0.5eV (continuous line) and W = 1eV (dashed line). The other parameters are: ω_c = 0.075 eV, U = 5.85 eV, and ε_f = –0.095 eV.

The important conclusion which follows from the behavior shown in Fig. 2 is that the collective mode is suppressed by hybridization.

4. POLARON GREEN'S FUNCTIONS

We denote by Λ the normal correlation function and by Y and $\bar{Y}$ the anomalous quantities. Statistical averages of a product of interaction operators can be divided in three groups: one is an average of products of conduction electron operators b_σ, b_σ^+ which is carried out with the help of the standard Wick theorem. The second group is composed of products of localized electron operators f_σ, f_σ^+ and such averages of correlated electrons require the generalized Wick theorem [15]. The third group of operators is formed by phonon cloud operators and for this group it is necessary to use a special relation (10).

Diagrammatic equations for polaron Green's functions of impurity electrons are represented in Fig. 3.

The general form includes the anomalous zero-th order Green's functions of conduction electrons F^0 and $\bar{F}^0$, which vanish in the normal state:

$$\begin{aligned} g_{p\,\sigma\sigma'}(i\omega) &= \Lambda_{\sigma\sigma'}(i\omega) + \Lambda_{\sigma\sigma_1}(i\omega)G^0_{\sigma_1}(i\omega)g_{p\,\sigma_1\sigma'}(i\omega) - \Lambda_{\sigma\sigma_1}(i\omega)F^0_{\sigma_1,-\sigma_1}(i\omega)\bar{f}_{p\bar{\sigma}_1\sigma'}(i\omega) - \\ &\quad -Y_{\sigma\bar{\sigma}_1}(i\omega)G^0_{\bar{\sigma}_1}(-i\omega)\bar{f}_{p\bar{\sigma}_1\sigma'}(i\omega) - Y_{\sigma\bar{\sigma}_1}(i\omega)\bar{F}^0_{\bar{\sigma}_1,\sigma_1}(i\omega)g_{p\,\sigma_1\sigma'}(i\omega), \\ \bar{f}_{p\bar{\sigma}\sigma'}(i\omega) &= \bar{Y}_{\bar{\sigma}\sigma'}(i\omega) + \bar{Y}_{\bar{\sigma}\sigma_1}(i\omega)G^0_{\sigma_1}(i\omega)g_{p\,\sigma_1\sigma'}(i\omega) - \bar{Y}_{\bar{\sigma}\sigma_1}(i\omega)F^0_{\sigma_1,-\sigma_1}(i\omega)\bar{f}_{p,-\bar{\sigma}_1,\sigma'}(i\omega) + \\ &\quad +\Lambda_{\bar{\sigma}_1\bar{\sigma}}(-i\omega)G^0_{\bar{\sigma}_1}(-i\omega)\bar{f}_{p\bar{\sigma}_1\sigma'}(i\omega) + \Lambda_{\bar{\sigma}_1\bar{\sigma}}(-i\omega)\bar{F}^0_{\bar{\sigma}_1,\sigma_1}(i\omega)g_{p\,\sigma_1\sigma'}(i\omega). \end{aligned} \tag{19}$$

The examples of normal and anomalous correlation function are shown in Fig. 4. Solution of these equations has the form:

$$\begin{aligned} g_{p\sigma}(i\omega) &= \frac{1}{d_{p\sigma}(i\omega)}\Big[\Lambda_{p\sigma}(i\omega) - G^0_{\bar{\sigma}}(-i\omega)\Big(\Lambda_{p\sigma}(i\omega)\Lambda_{p\bar{\sigma}}(-i\omega) + Y_{p\sigma\bar{\sigma}}(i\omega)\bar{Y}_{p\bar{\sigma}\sigma}(i\omega)\Big)\Big], \\ \bar{f}_{p\bar{\sigma}\sigma}(i\omega) &= \frac{1}{d_{p\sigma}(i\omega)}\Big[\bar{Y}_{p\bar{\sigma}\sigma}(i\omega) + F^0_{\bar{\sigma}\sigma}(i\omega)\Big(\Lambda_{p\sigma}(i\omega)\Lambda_{p\bar{\sigma}}(-i\omega) + Y_{p\sigma\bar{\sigma}}(i\omega)\bar{Y}_{p\bar{\sigma}\sigma}(i\omega)\Big)\Big], \\ d_{p\sigma}(i\omega) &= 1 - \Lambda_{p\sigma}(i\omega)G^0_{\sigma}(i\omega) - \Lambda_{p\bar{\sigma}}(-i\omega)G^0_{\bar{\sigma}}(-i\omega) + \bar{Y}_{p\bar{\sigma}\sigma}(i\omega)F^0_{\sigma\bar{\sigma}}(i\omega) + \bar{Y}_{p\sigma\bar{\sigma}}(i\omega)F^0_{\bar{\sigma}\sigma}(i\omega) + \\ &\quad +\Big[G^0_{\sigma}(i\omega)G^{(0)}_{\bar{\sigma}}(-i\omega) + F^0_{\sigma\bar{\sigma}}(i\omega)\bar{F}^0_{\bar{\sigma}\sigma}(i\omega)\Big]\Big[\Lambda_{p\sigma}(i\omega)\Lambda^{(0)}_{p\bar{\sigma}}(-i\omega) + Y_{p\sigma\bar{\sigma}}(i\omega)\bar{Y}_{p\bar{\sigma}\sigma}(i\omega)\Big]. \end{aligned} \tag{20}$$

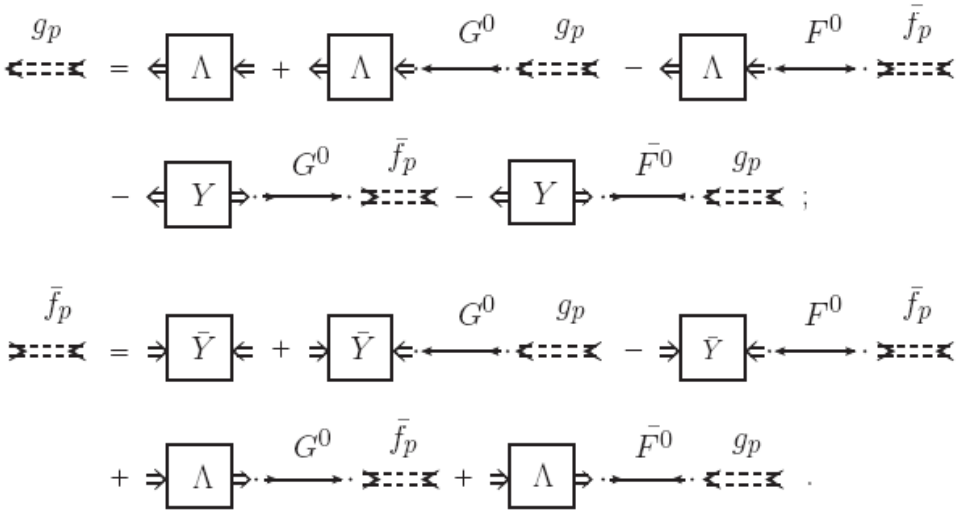

Fig. 3 – Dyson type equations for full Green's functions of impurity electrons.

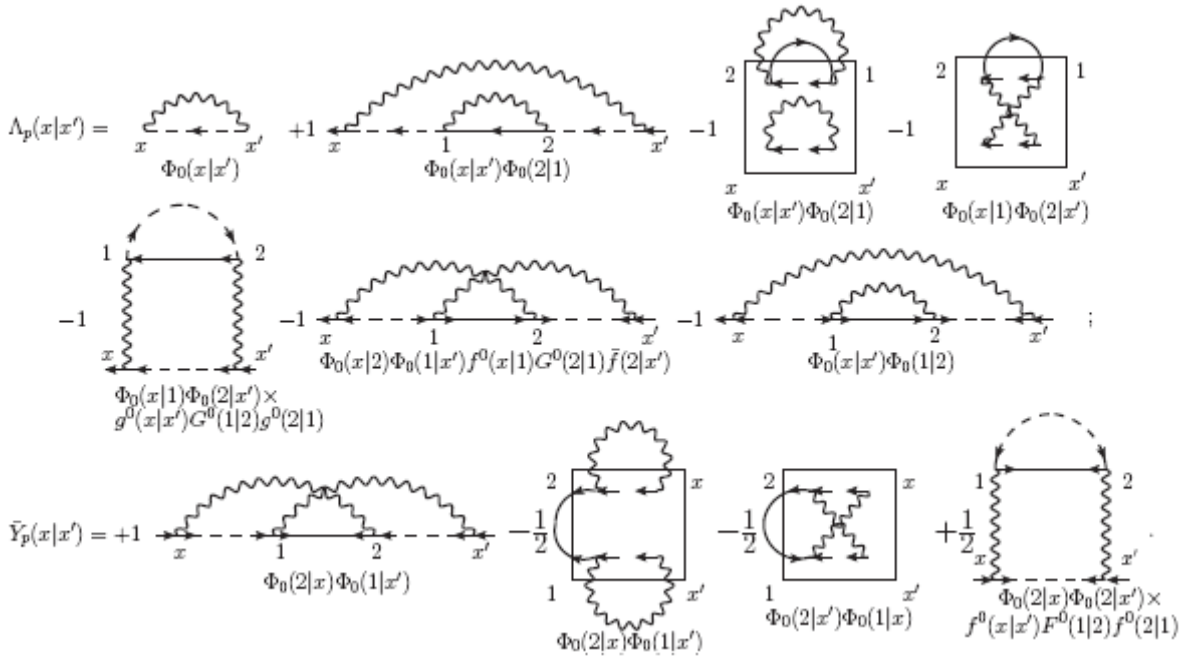

Fig. 4 – The examples of correlation functions in first order of perturbation theory. The rectangles depict $G_2^{(irr)}$.

The system of equations for the renormalized Green's functions G, F, and $\bar{F}$ of conduction electrons can be obtained in a similar way. Their diagrammatic representation is shown in Fig. 5.

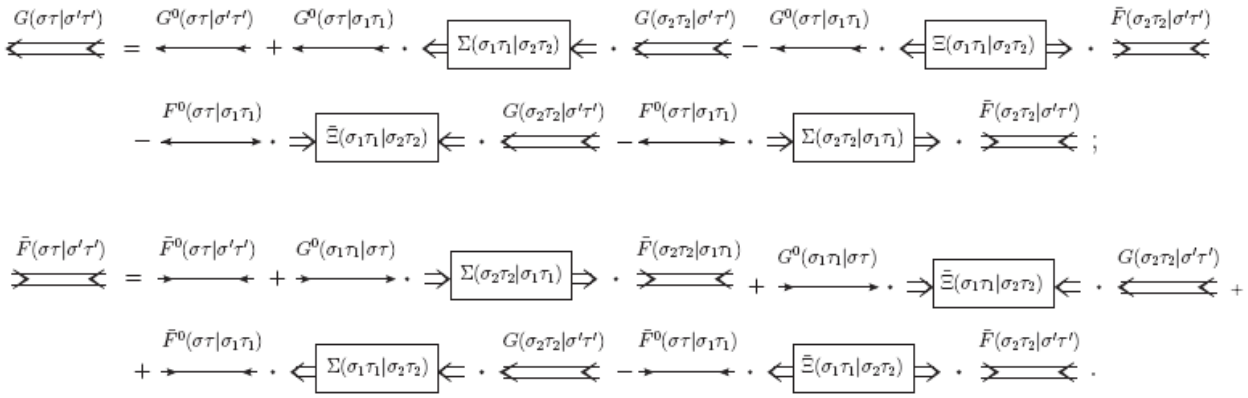

Fig. 5 – Renormalized Green's functions of the conduction electrons. Thin solid lines are zero-order functions. The double thin lines represent the renormalized functions and Σ, Ξ, and $\bar{\Xi}$ are the mass operators of the conduction electrons.

Solutions of the whole set of equations are determined by the normal Σ and by the two anomalous Ξ and $\bar{\Xi}$ mass operators, which we identify below as correlation functions of localized electrons:

$$\Sigma(\sigma\tau,\sigma'\tau') = \Lambda(\sigma\tau,\sigma'\tau'), \Xi(\sigma\tau,\sigma'\tau') = Y(\sigma\tau,\sigma'\tau'), \bar{\Xi}(\sigma\tau,\sigma'\tau') = \bar{Y}(\sigma\tau,\sigma'\tau'). \tag{21}$$

Thus, the correlation functions Λ, Y, and $\bar{Y}$ of the Dyson type equation for localized electrons coincide with the mass operators of the Dyson equations for conduction electrons.

5. CONCLUSIONS

We have developed a diagrammatic approach for the Anderson-Holstein model in the case of strong Coulomb intra-atomic interaction of impurity electrons and strong electron-phonon interaction of the

impurity electrons with optical phonons. Both interactions are taken into account as the main part of the Hamiltonian.

Dynamics of the phonon clouds of polarons has been investigated and the renormalization of their collective frequency has been described in detail. We have proved that at zero temperature hybridization causes a continuous softening of the collective mode.

We have formulated the system of Dyson-type equations which determines the relation between the full propagators of impurity electrons and their correlation functions. We have also found the system of Dyson equations for the full Green's functions of conduction electrons and for their mass operators. As a consequence of this analysis we have proven the identity between the impurity correlation functions and the mass operators of the conduction electrons.

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