Optical conductivity of a strong-coupling polaron

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Abstract

The polaron optical conductivity is derived within the strong-coupling expansion, which is asymptotically exact in the strong-coupling limit. The polaron optical conductivity band is provided by the multiphonon optical transitions. The polaron optical conductivity spectra calculated within our analytic strong-coupling approach and the numerically accurate Diagrammatic Quantum Monte Carlo (DQMC) data are in a good agreement with each other at large $\alpha \gtrsim 9$.

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I. INTRODUCTION

The optical conductivity of the Fröhlich polaron model attracted attention for years \[1\]. In the regime of weak coupling, the optical absorption of a polaron was calculated using different methods, e. g., Green’s function method \[2\], the Low-Lee-Pines formalism \[3, 4\], perturbation expansion of the current-current correlation function \[5\]. The strong-coupling polaron optical conductivity was calculated taking into account one-phonon \[6\] and two-phonon \[7\] transitions from the polaron ground state to the polaron relaxed excited state (RES). In fact the present work finalizes the project started in Ref. \[6\]. Using the path integral response formalism, the impedance function of an all-coupling polaron was calculated by FHIP \[9\] on the basis of the Feynman polaron model \[10\]. Developing further the FHIP approach, the optical conductivity was calculated in the path-integral formalism at zero temperature \[8\] and at finite temperatures \[12\]. In Ref. \[16\], the extension of the method of Ref. \[8\] accounting for the polaron damping (for the polaron coupling constant \[\alpha \lesssim 8\]) and the asymptotic strong-coupling approach using the Franck-Condon (FC) picture for the optical conductivity (for \[\alpha \gtrsim 8\]) have given reasonable results for the polaron optical conductivity at all values of \[\alpha\]. The concept of the RES and FC polaron states played a key role in the understanding of the mechanism of the polaron optical conductivity \[6–8, 11–13\].

Recently, the Diagrammatic Quantum Monte Carlo (DQMC) numerical method has been developed \[14, 15\], which provides accurate results for the polaron characteristics in all coupling regimes. The analytic treatment \[8\] was intended to be valid at all coupling strengths. However, it is established in \[7, 8, 11\] that the linewidth of the obtained spectra \[8\] is unreliable for \[\alpha \gtrsim 7\]. Nevertheless, the position of the peak attributed to RES in Ref. \[8\] is close to the maximum of the polaron optical conductivity band calculated using DQMC up to very large values of \[\alpha\] (see Fig. 1).

An extension of the path-integral approach \[8\] performed in Ref. \[16\] gives a good agreement with DQMC for weak and intermediate coupling strengths. In the strong-coupling limit, in Ref. \[16\] the adiabatic strong-coupling expansion was applied. That expansion, however, is not exact in the strong-coupling limit because of a parabolic approximation \[17\] for the adiabatic potential.

In the present work, the strong-coupling approach of Ref. \[16\] is extended in order to obtain the polaron optical conductivity which is asymptotically exact in the strong-coupling
We develop the multiphonon strong-coupling expansion using numerically accurate in the strong-coupling limit polaron energies and wave functions and accounting for non-adiabaticity.

II. OPTICAL CONDUCTIVITY

We consider the electron-phonon system with the Hamiltonian written down in the Feynman units \((\hbar = 1\), the carrier band mass \(m_b = 1\), and the LO-phonon frequency \(\omega_{LO} = 1\))

\[
H = \frac{p^2}{2} + \sum_q \left( b_q^+ b_q + \frac{1}{2} \right) + \frac{1}{\sqrt{V}} \sum_q \frac{\sqrt{2} \sqrt{2 \pi \alpha}}{q} (b_q + b_q^+) e^{i q \cdot r}.
\]

(1)

where \(r, p\) represent the position and momentum of an electron, \(b_q^+, b_q\) denote the creation and annihilation operators for longitudinal optical (LO) phonons with wave vector \(q\), and \(V_q\) describes the amplitude of the interaction between the electrons and the phonons. For the Fröhlich electron-phonon interaction, the amplitude of the electron – LO-phonon interaction is

\[
V_q = \frac{1}{\sqrt{V}} \frac{\sqrt{2} \sqrt{2 \pi \alpha}}{q}
\]

(2)

with the crystal volume \(V\), and the electron-phonon coupling constant \(\alpha\).

The polaron optical conductivity describes the response of the system with the Hamiltonian (1) to an applied electromagnetic field (along the \(z\)-axis) with frequency \(\omega\). This optical response is expressed using the Kubo formula with a dipole-dipole correlation function:

\[
\text{Re} \sigma (\omega) = \frac{n_0 \omega}{2} \left( 1 - e^{-\beta \omega} \right) \int_{-\infty}^{\infty} e^{i \omega t} \langle d_z (t) d_z \rangle \ dt,
\]

(3)

where \(d = -e_0 r\) is the electric dipole moment, \(e_0\) is the unit charge, \(\beta = \frac{1}{k_b T}\), \(n_0\) is the electron density. In the zero-temperature limit, the optical conductivity (3) measured in units of \(e_0^2\) becomes

\[
\text{Re} \sigma (\omega) = \frac{\omega}{2} \int_{-\infty}^{\infty} e^{i \omega t} f_{zz} (t) \ dt,
\]

(4)

with the correlation function

\[
f_{zz} (t) \equiv \langle z (t) z (0) \rangle = \langle \Psi_0 | e^{i H z} e^{-i H z} | \Psi_0 \rangle,
\]

(5)

where \(|\Psi_0\rangle\) is the ground-state wave function of the electron-phonon system.
Within the strong-coupling approach, the ground-state wave function is chosen as the product of a trial wave function of an electron $|\psi_0^{(e)}\rangle$ and of a trial wave function of a phonon subsystem $|\Phi_{ph}\rangle$:

$$|\Psi_0\rangle = |\psi_0^{(e)}\rangle |\Phi_{ph}\rangle.$$  \hspace{1cm} (6)

The phonon trial wave function is written as the strong-coupling unitary transformation applied to the phonon vacuum $|\Phi_{ph}\rangle = U |0_{ph}\rangle$.

$$|\Phi_{ph}\rangle = U |0_{ph}\rangle.$$  \hspace{1cm} (7)

with the unitary operator

$$U = e^{\sum_q (f_q b_q - f_q^* b_q^+)}.$$  \hspace{1cm} (8)

and the variational parameters $\{f_q\}$. The transformed Hamiltonian $\tilde{H} \equiv U^{-1} H U$ takes the form

$$\tilde{H} = \tilde{H}_0 + W$$  \hspace{1cm} (9)

with the terms

$$\tilde{H}_0 = \frac{p^2}{2} + \sum_q |f_q|^2 + V_a (r) + \sum_q \left( b_q^+ b_q + \frac{1}{2} \right),$$  \hspace{1cm} (10)

$$W = \sum_q \left( W_q b_q + W_q^* b_q^+ \right).$$  \hspace{1cm} (11)

Here, $W_q$ are the amplitudes of the renormalized electron-phonon interaction

$$W_q = \sqrt{\frac{2 \sqrt{2 \pi \alpha}}{q \sqrt{V}}} \left( e^{i q \cdot r} - \rho_q \right),$$  \hspace{1cm} (12)

where $\rho_q$ is the expectation value of the operator $e^{i q \cdot r}$ with the trial electron wave function $|\psi_0^{(e)}\rangle$:

$$\rho_q = \langle \psi_0^{(e)} | e^{i q \cdot r} | \psi_0^{(e)} \rangle,$$  \hspace{1cm} (13)

and $V_a (r)$ is the self-consistent potential energy for the electron,

$$V_a (r) = - \sum_q \frac{4 \sqrt{2 \pi \alpha}}{q^2 V} \rho_{-q} e^{i q \cdot r}.$$  \hspace{1cm} (14)

Averaging the Hamiltonian (9) with the phonon vacuum $|0\rangle$ and with the trial electron wave function $|\psi_0\rangle$, we arrive at the following variational expression for the ground-state
energy

\[ E_0 = \langle \Psi_0 | H | \Psi_0 \rangle = \left\langle \psi_0 \left| \frac{P^2}{2} \right| \psi_0 \right\rangle + \sum_q |f_q|^2 \]

\[ - \sum_q \left( V_q f_q^* \rho_q + V_q^* f_q \rho_{-q} \right), \quad (15) \]

After minimization of the polaron ground-state energy (15), the parameters \( f_q \) acquire their optimal values

\[ f_q = V_q \rho_q. \quad (16) \]

The ground-state energy with \( \{ f_q \} \) given by Eq. (16) takes the form

\[ E_0 = \left\langle \psi_0 \left| \frac{P^2}{2} \right| \psi_0 \right\rangle - \sum_q |V_q|^2 |\rho_q|^2. \quad (17) \]

With the strong-coupling Ansatz (6) for the polaron ground-state wave function and after the application of the unitary transformation (8), the correlation function (5) takes the form

\[ f_{zz}(t) = \left\langle 0_{ph} \left| \left\langle \psi_0 \right| e^{itH} z e^{-itH} z \right| \psi_0 \right\rangle \left| 0_{ph} \right\rangle. \quad (18) \]

This correlation function can be expanded using a complete orthogonal set of intermediate states \( |j \rangle \) and the completeness property:

\[ \sum_j |j \rangle \langle j| = 1. \quad (19) \]

In the present work, we use the intermediate basis of the Franck-Condon (FC) states. The FC states correspond to the equilibrium phonon configuration for the ground state. Thus the FC wave functions are the exact eigenstates of the Hamiltonian \( \tilde{H}_0 \). Further on, the FC wave functions are written in the spherical-wave representation as \( |\psi_{n,l,m} \rangle = R_{n,l}(r) Y_{l,m}(\theta, \varphi) \) where \( R_{n,l}(r) \) are the radial wave functions, and \( Y_{l,m}(\theta, \varphi) \) are the spherical harmonics, \( l \) is the quantum number of the angular momentum, \( m \) is the \( z \)-projection of the angular momentum, and \( n \) is the radial quantum number\(^1\). The energy levels for the eigenstates of the Hamiltonian \( \tilde{H}_0 \) are denoted \( E_{n,l} \).

\(^1\) In this classification, the ground-state wave function is \( |\psi_{0,0,0} \rangle \equiv |\psi_0 \rangle \).
Using (19) with that complete and orthogonal basis, we transform (18) to the expression

\[
f_{zz}(t) = \sum_{n,l,m,n',l',m'} \langle \psi_{n',l',m'} | z | \psi_{n',l',m'} \rangle \langle \psi_{n,l,m} | z | \psi_0 \rangle \\
\times \left( \left| 0_{ph} \right| e^{i\tilde{H}t} \left| \psi_{n,l,m} \right\rangle \left\langle \psi_{n',l',m'} | e^{-i\tilde{H}t} | \psi_{n',l',m'} \right\rangle \left| 0_{ph} \right\rangle \right) .
\]  

(20)

So far, the only approximation made in (20) is the strong-coupling Ansatz for the polaron ground-state wave function. However, in order to obtain a numerically tractable expression for the polaron optical conductivity, an additional approximation valid in the strong-coupling limit must be applied to the matrix elements of the evolution operator \( e^{-it\tilde{H}} \) with the Hamiltonian of the electron-phonon system \( \tilde{H} \) given by formula (9). According to Ref. [18], in the strong-coupling limit, the matrix elements of the Hamiltonian of the electron-phonon system between states corresponding to different energy levels are of order of magnitude \( \alpha^{-4} \). Therefore in the strong-coupling regime these matrix elements can be neglected; this is called the adiabatic or the Born-Oppenheimer (BO) approximation [18], because of its strict analogy with the Born-Oppenheimer adiabatic approximation in the theory of molecules and crystals [19], p. 171). Consequently, in the further treatment we neglect the matrix elements \( \left\langle \psi_{n,l,m} | e^{-i\tilde{H}t} | \psi_{n',l',m'} \right\rangle \) for the FC states with different energies, \( E_{n,l} \neq E_{n',l'} \). The same scheme was used in the theory of the multi-phonon optical processes for bound electrons interacting with phonons [20, 21].

Strictly speaking, the summation over the excited polaron states in Eq. (20) must involve the transitions to both the discrete and continuous parts of the polaron spectrum. A transition to the states of the continuous spectrum means that the electron leaves the polaron potential well. Therefore these transitions can be attributed to the “polaron dissociation”. The transitions to the continuous spectrum are definitely beyond the adiabatic approximation. As shown in Ref. [21], the transition probability to the states of the continuous spectrum is very small compared with the transition probability between the ground and the first excited state (which belongs to the discrete part of the polaron energy spectrum). We neglect here the contribution to the polaron optical conductivity due to the transitions to the continuous spectrum.

The matrix elements neglected within the adiabatic approximation correspond to the transitions between FC states with different energies due to the electron-phonon interaction. Hence these transitions can be called non-adiabatic. The adiabatic approximation is
related to the matrix elements of the evolution operator $e^{-it\tilde{H}}$. On the contrary, the matrix elements of the transitions between different FC states for the electric dipole moment are, in general, not equal to zero. Moreover, these transitions can be accompanied by the emission of phonons. The electron FC wave functions constitute a complete orthogonal set. However, the corresponding phonon wave functions can be non-orthogonal because of a different shift of phonon coordinates for different electron states. This makes multi-phonon transitions possible. It is important to note that in our treatment we neglect only the non-adiabatic transitions between the electron states with different energies. On the contrary, the transitions within one and the same degenerate level can be non-adiabatic. This internal non-adiabaticity (i.e., the non-adiabaticity of the transitions within one and the same degenerate level) is taken into account in the subsequent treatment.

It is useful to stress the difference between the strong-coupling Ansatz and the adiabatic approximation. The strong-coupling Ansatz consists of the choice of the trial variational ground state wave function for the electron-phonon system in the factorized form \([3]\). The adiabatic approximation means neglecting the matrix elements of the evolution operator between internal polaron states with different energies. These two approximations are not the same, but they both are valid in the strong-coupling regime and consistent with each other.

The correlation function \([20]\) is transformed in the following way. The exponents $e^{it\tilde{H}}$ and $e^{-it\tilde{H}}$ are disentangled:

$$e^{-it\tilde{H}} = e^{-it\tilde{H}_0} T \exp \left( -i \int_0^t ds W(s) \right), \quad (21)$$

$$e^{it\tilde{H}} = e^{it\tilde{H}_0} T \exp \left( i \int_0^t ds W(-s) \right) \quad (22)$$

where $W(s)$ is the renormalized electron-phonon interaction Hamiltonian $\tilde{W}$ in the interaction representation,

$$W(s) \equiv e^{is\tilde{H}_0} W e^{-is\tilde{H}_0}. \quad (23)$$
This gives us the result

\[
f_{zz}(t) = \sum_{n,l,m, n',l',m', n'',l'',m''} \langle \psi_{n,l,m} | z | \psi_{n',l',m'} \rangle \langle \psi_{n',l',m'} | z | \psi_{0} \rangle e^{it(E_{0}-E_{n',l',m'})} \\
\times \left\langle 0_{ph} \left| \psi_{0} \right| \text{Tr} \left[ T \exp \left( i \int_{0}^{t} dsW(s) \right) \right] \left| 0_{ph} \right\rangle \right. \\
\times \left\langle \psi_{n'',l'',m''} \left| \psi_{n',l',m'} \right| \right. \\
\times \left\langle \psi_{n',l',m'} \left| T \exp \left( -i \int_{0}^{t} dsW(s) \right) \right| 0_{ph} \right\rangle.
\]  

(24)

Within the adiabatic approximation, the optical conductivity is simplified. The full details of the derivation are described in the Appendix A. First, using the selection rules for the dipole matrix elements, the spherical symmetry of the Hamiltonian \( \tilde{H} \) and the adiabatic approximation, the correlation function (24) is reduced to the form

\[
f_{zz}(t) = \sum_{n} D_{n} e^{-i\Omega_{n,0}t} \\
\times \left\langle \psi_{n,1,0} \left| 0_{ph} \right| \text{Tr} \left[ T \exp \left( -i \int_{0}^{t} dsW(s) \right) \right] \left| 0_{ph} \right\rangle \right. \\
\times \left\langle \psi_{n',l',m'} \left| \psi_{n,1,0} \right| \right. \\
\times \left\langle \psi_{n,1,0} \left| \psi_{n',l',m'} \right| \right. \\
\times \left\langle \psi_{n',l',m'} \left| T \exp \left( -i \int_{0}^{t} dsW(s) \right) \right| 0_{ph} \right\rangle. 
\]

(25)

where \( \Omega_{n,0} \) is the FC transition frequency

\[
\Omega_{n,0} \equiv E_{n,1} - E_{0}, 
\]

(26)

and \( D_{n} \) is the squared modulus of the dipole transition matrix element

\[
D_{n} = |\langle \psi_{0} | z | \psi_{n,1,0} \rangle|^{2}. 
\]

(27)

Within the adiabatic approximation, the partial (with the electron wave functions) averaging of the operator T-exponent in (25) can be exactly performed (see details in Appendix A). As a result, the optical conductivity is transformed to the expression

\[
\text{Re} \sigma(\omega) = \frac{\omega}{6} \sum_{n} D_{n} \int_{-\infty}^{\infty} e^{i(\omega-\Omega_{n,0})t} \\
\times \left\langle 0_{ph} \left| \text{Tr} \left( T \exp \left[ -i \int_{0}^{t} dsW^{(n)}(s) \right] \right) \right| 0_{ph} \right\rangle dt. 
\]

(28)

The T-exponent in (28) contains the finite-dimensional matrix \( W^{(n)}(s) \) depending on the phonon coordinates:

\[
\left( W_{k,l,m}^{(n)} \right)_{m_{1},m_{2}} = \langle \psi_{n,1,m_{1}} | W_{k,l,m} | \psi_{n,1,m_{2}} \rangle 
\]

(29)
where $W_{k,l,m}$ are the amplitudes of the electron-phonon interaction in the basis of spherical wave functions.

Because the kinetic energy of the phonons is of order $\alpha^{-4}$ compared to the leading term of the Hamiltonian [18], we neglect this kinetic energy in the present work, because the treatment is related to the strong-coupling regime. As a result, $Q_{k,l,m}$ commute with the Hamiltonian $\tilde{H}_0$, so that in (28), $\mathbb{W}^{(n)}(s) = \mathbb{W}^{(n)}$. Furthermore, in a finite-dimensional basis $\{|\psi_{n,l,m}\rangle\}$ for a given level $(n,l)$, all eigenvalues of the Hamiltonian $\tilde{H}_0$ are the same. Therefore the T-exponent entering (28) in that finite-dimensional basis turns into a usual exponent. As a result, the strong-coupling polaron optical conductivity (28) takes the form

$$\text{Re} \sigma(\omega) = \frac{\omega}{6} \sum_n D_n \int_{-\infty}^{\infty} e^{i(\omega - \Omega_{n,0})t} \langle 0_{ph} | \text{Tr} \exp(-i\mathbb{W}^{(n)}t) | 0_{ph} \rangle dt. \tag{30}$$

The matrix interaction Hamiltonian (29) depends on the phonon coordinates, and the matrices $\mathbb{W}^{(n)}_{k,l,m}$ with different $m$ for one and the same degenerate energy level do not commute with each other. According to the Jahn – Teller theorem [24], for a degenerate level there does not exist a unitary transformation which simultaneously diagonalizes all matrices $\mathbb{W}^{(n)}_{k,l,m}$ in a basis that does not depend on the phonon coordinates. The manifestations of that theorem are attributed to the Jahn – Teller effect. Therefore, because we neglect the non-commutation of the matrices $\mathbb{W}^{(n)}_{k,l,m}$, the Jahn – Teller effect is omitted.

In fact, neglecting the Jahn – Teller effect is not necessary. The averaging in Eq. (30) is performed exactly using the effective phonon modes similarly to Ref. [26] (see the details in Appendix B). As a result, we arrive at the following expression for the strong-coupling polaron optical conductivity

$$\text{Re} \sigma(\omega) = \frac{\omega}{3\pi^2} \sum_n \frac{D_n}{a_0^{(n)}} \int_{-\infty}^{\infty} dx_0 \int_{-\infty}^{\infty} dx_1 \int_{-\infty}^{\infty} dx_2 \int_{-\infty}^{\infty} dy_1 \int_{-\infty}^{\infty} dy_2 \times \sum_{j=1}^{3} \exp \left\{-\frac{1}{2} \left[ \frac{1}{2} \sum_{m=1,2} \left( x_m^2 + y_m^2 \right) + \frac{\omega - \Omega_{n,0} - \frac{a_2^{(n)}}{2\sqrt{5}\pi} \lambda_j(Q_2)^2}{\left( a_0^{(n)} \right)^2} \right] \right\}. \tag{31}$$

Here, $\lambda_j(Q_2)$ are the eigenvalues for the matrix interaction Hamiltonian, which are explicitly determined in the Appendix B by the formula (B20). The coefficients $a_0^{(n)}$ and $a_2^{(n)}$ are given by (B11) and (B12), respectively. The polaron optical conductivity given by the expression (31), is in fact an envelope of the multiphonon polaron optical conductivity band with
the correlation function (28) provided by the phonon-assisted transitions from the polaron ground state to the polaron RES. This result is consistent with Ref. [6], where the same paradigm of the phonon-assisted transitions to the polaron RES was exploited, but the calculation was limited to the one-phonon transition.

In order to reveal the significance of the Jahn–Teller effect for the polaron, we alternatively calculate

\[
\langle 0_{ph} | \text{Tr} \exp \left( -i \mathcal{W}^{(n)} t \right) | 0_{ph} \rangle
\]

neglecting the non-commutation of the matrices \( \mathcal{W}^{(n)} \), as described in the Appendix B. 2. The resulting expression for the polaron optical conductivity is much simpler than formula (31) and is similar to the expression (3) of Ref. [16]:

\[
\text{Re} \sigma (\omega) = \omega \sum_n \sqrt{\frac{\pi}{2 \omega^{(n)}_s}} D_n \exp \left( -\frac{\left( \omega - \Omega_{n,0} \right)^2}{2 \omega^{(n)}_s} \right),
\]

with the parameter (often called the Huang-Rhys factor)

\[
\omega^{(n)}_s = \frac{1}{2} \left( a^{(n)}_0 \right)^2 + \frac{1}{4\pi} \left( a^{(n)}_2 \right)^2.
\]

The strong-coupling electron energies and wave functions in Eq. (28) can be calculated using different approximations. For example, within the Landau-Pekar (LP) approximation [17], the trial wave function \( |\psi_0\rangle \) is chosen as the ground state of a 3D oscillator. Within the Pekar approximation [21], \( |\psi_0\rangle \) is chosen in the form

\[
|\psi_0 (r)\rangle = Ce^{-ar} \left( 1 + ar + br^2 \right)
\]

with the variational parameters \( a \) and \( b \). Finally, the trial ground state wave function can be determined numerically exactly following Miyake [22] (see also [23], Chap. 5.22). Within the LP approximation, formula (32) reproduces the polaron optical conductivity obtained in Ref. [16].

In the LP approximation, the matrix elements \( \langle \psi_0 | z | \psi_{n,1,0} \rangle \) are different from zero only for \( n = 1 \), i. e. only for the \( 1s \rightarrow 2p \) transition. Beyond the LP approximation, also the transitions to other excited states are allowed because of the nonparabolicity of the self-consistent potential \( V_a (r) \). The use of exact strong-coupling wave functions, instead of the LP wave functions, may significantly influence the optical conductivity. In the present treatment we use the numerically exact electron energies and wave functions of both ground and first excited states according to Ref. [22]. The FC transition energies \( \Omega_{n,0} \) to leading order of the strong-coupling approximation are determined according to (26). In order to
account for the corrections of the FC energy with accuracy up to $\alpha^0$, we add to $\Omega_{n,0}$ the correction $\Delta\Omega_{\text{FC}} \approx -3.8$ from Ref. [16]. Because we use the numerically accurate strong-coupling wave functions and energies corresponding to Miyake [22], the formula (24) is asymptotically exact in the strong-coupling limit, at least in its leading term in powers of $\alpha^{-2}$.

III. RESULTS AND DISCUSSION

In Figs. 2 to 3, we have plotted the polaron optical conductivity spectra calculated for different values of the coupling constant $\alpha$. The optical conductivity spectra calculated within the present strong-coupling approach taking into account the Jahn – Teller effect are shown by the solid curves. The optical conductivity derived neglecting the Jahn – Teller effect is shown by the dashed curves. It is worth mentioning that there is little difference in the optical conductivity spectra between those calculated with and without the Jahn – Teller effect. The optical conductivity obtained in Ref. [16] with the Landau-Pekar (LP) adiabatic approximation is plotted with dash-dotted curves. The full dots show the numerical Diagrammatic Quantum Monte Carlo (DQMC) data [15, 16]. The FC transition frequency for the transition to the first excited FC state $\Omega_{1,0} \equiv \Omega_{\text{FC}}$ and the RES transition frequency $\Omega_{\text{RES}}$ are explicitly indicated in the figures.

The polaron optical conductivity spectra calculated within the present strong-coupling approach are shifted to lower frequencies with respect to the optical conductivity spectra calculated within the LP approximation of Ref. [16]. This shift is due to the use of the numerically accurate strong coupling energy levels and wave functions of the internal polaron states, and of the numerically accurate self-consistent adiabatic polaron potential.

According to the selection rules for the matrix elements of the electron-phonon interaction, there is a contribution to the polaron optical conductivity from the phonon modes with angular momentum $l = 0$ ($s$-phonons) and with angular momentum $l = 2$ ($d$-phonons). The $s$-phonons are fully symmetric, therefore they do not contribute to the Jahn – Teller effect, while the $d$-phonons are active in the Jahn – Teller effect. The contribution of the $d$-phonons to the optical conductivity spectra is not small compared to the contribution of the $s$-phonons. However, the distinction between the optical conductivity spectra calculated with and without the Jahn – Teller effect is relatively small.
For $\alpha = 8$ and $\alpha = 8.5$, the maxima of the polaron optical conductivity spectra, calculated within the present strong-coupling approach are positioned to the low frequency side of the maxima of those calculated using the DQMC method. The agreement between our strong-coupling polaron optical conductivity spectra and the numerical DQMC data improves with increasing alpha. This is in accordance with the fact that the present strong-coupling approach for the polaron optical conductivity is asymptotically exact in the strong-coupling limit.

The total polaron optical conductivity must satisfy the sum rule \[ \int_0^\infty \text{Re} \sigma (\omega) \, d\omega = \frac{\pi}{2}. \] (35)

In the weak- and intermediate-coupling regimes at $T = 0$, there are two contributions to the left-hand side of that sum rule: (1) the contribution from the polaron optical conductivity for $\omega > \omega_{LO}$ and (2) the contribution from the “central peak” at $\omega = 0$, which is proportional to the inverse polaron mass \[ 25 \]. In the asymptotic strong-coupling regime, the inverse to the polaron mass is of order $\alpha^{-4}$, and hence the contribution from the “central peak” to the polaron optical conductivity is beyond the accuracy of the present approximation (where we keep the terms $\propto \alpha^{-2}$ and $\propto \alpha^0$).

As discussed above, in the present work the transitions from the ground state to the states of the continuous part of the polaron energy spectrum are neglected. Therefore the integral over the frequency [the left-hand side of (35)] for the optical conductivity calculated within the present strong-coupling approximation can be (relatively slightly) smaller than $\pi/2$. The relative contribution of the transitions to the continuous part of the polaron spectrum, $\Delta_c$, can be therefore estimated as

$$ \Delta_c \equiv 1 - \frac{2}{\pi} \int_0^\infty \text{Re} \sigma (\omega) \, d\omega, $$

where the right-hand side is obtained by a numerical integration of $\text{Re} \sigma (\omega)$ calculated within the present strong-coupling approach. This numeric estimation shows that for $\alpha > 8$, $\Delta_c < 0.01$. Moreover, with increasing $\alpha$, the relative contribution of the transitions to the continuous part of the polaron spectrum falls down. This confirms the accuracy of the present strong-coupling approach.

In Refs. \[ 27, 28 \], the optical conductivity of a strong-coupling polaron was calculated assuming that in the strong-coupling regime the polaron optical response is provided mainly
by the transitions to the continuous part of the spectrum (these transitions are called there “the polaron dissociation”). This concept is in contradiction both with the early estimation by Pekar [21] discussed above and with the very small weight of those transitions shown in Fig. 4. The approach of Ref. [27] in fact takes into account only a small part of the strong-coupling polaron optical conductivity – namely, the high-frequency “tail” of the optical conductivity spectrum.

When comparing the polaron optical conductivity spectra calculated in the present work with the DQMC data [15, 16], we can see that the present approach, with respect to DQMC, underestimates the high-frequency part of the polaron optical conductivity. This difference, however, gradually diminishes with increasing \( \alpha \), in accordance with the fact that the present method is an asymptotic strong-coupling approximation.

Because the optical conductivity spectra calculated in the present strong-coupling approximation using the expressions (31) and (32) represent the envelopes of the RES peak with the multi-phonon satellites, the separate peaks are not explicitly seen in those spectra. The FC and RES peaks are indicated in the figures by the arrows. The FC transition frequency \( \Omega_{1,0} \) in the strong-coupling case is positioned close to the maximum of the polaron optical conductivity band (both calculated within the present approach and within DQMC). The RES transition frequency is positioned one \( \omega_{LO} \) below the onset of the LO-sidebands. Note that the strong-coupling polaron optical conductivity derived in Refs. [29] contains only the zero-phonon (RES) line and no phonon satellites at all. In contrast, in the present calculation, the maximum of the polaron optical conductivity spectrum shifts to higher frequencies with increasing \( \alpha \), so that the multiphonon processes invoking large number of phonons become more and more important, in accordance with predictions of Refs. [6, 8].

It is worth noting the following important point: the maximum of the polaron optical conductivity band can be hardly interpreted as a broadened transition to an FC state on the following reasons. Formula (28) describes a set of multi-phonon peaks. In the simplifying approximation which neglects the Jahn – Teller effect (see Ref. [16]), those peaks are positioned at the frequencies \( \omega = \tilde{\Omega}_{n,0} + k \), where \( k \) is the number of emitted phonons and is the frequency of the zero-phonon line. The frequencies \( \tilde{\Omega}_{n,0} \) do not coincide with the FC transition frequencies but are determined by

\[
\tilde{\Omega}_{n,0} = \Omega_{n,0} - \omega^{(n)},
\]  

(37)
where the Huang-Rhys factor $\omega_s^{(n)}$ describes the energy shift due to lattice relaxation. The physical meaning of the parameters $\omega_s^{(n)}$ obviously implies that the peaks at $\omega = \tilde{\Omega}_{n,0} + k$ should be attributed to transitions to the RES with emission of $k$ phonons. So, the so-called “FC transition” is realized as the envelope of a series of phonon sidebands of the polaron RES but not as a transition to the FC state. The account of the Jahn-Teller effects in general makes the multiphonon peak series non-equidistant, but it changes nothing in the concept of the internal polaron states which is discussed above.

IV. CONCLUSIONS

We have derived the polaron optical conductivity which is asymptotically exact in the strong-coupling limit. The strong-coupling polaron optical conductivity band is provided by the multiphonon transitions from the polaron ground state to the polaron RES and has the maximum positioned close to the FC transition frequency. With increasing the electron-phonon coupling constant $\alpha$, the polaron optical conductivity band shape gradually tends to that provided by the Diagrammatic Quantum Monte Carlo (DQMC) method. This agreement demonstrates the importance of the multiphonon processes for the polaron optical conductivity in the strong-coupling regime.

The obtained polaron optical conductivity with a high accuracy satisfies the sum rule \cite{25}, what gives us an evidence of the fact that in the strong-coupling regime the dominating contribution to the polaron optical conductivity is due to the transitions to the internal polaron states, while the contribution due to the transitions to the continuum states is negligibly small.

Accurate numerical results, obtained using DQMC method \cite{15}, – modulo the linewidths for sufficiently large $\alpha$ – and the analytically exact in the strong-coupling limit polaron optical conductivity of the present work, as well as the analytical approximation of Ref. \cite{16} confirm the essence of the mechanism for the optical absorption of Fröhlich polarons, which were proposed in Refs. \cite{8,11}.
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Appendix A: Correlation function

The dipole-dipole correlation function \( f_{zz}(t) \) given by (24) is further simplified within the adiabatic approximation and using the selection rules for the dipole transition matrix elements and the symmetry properties of the polaron Hamiltonian. First, according to the selection rules, the matrix element \( \langle \psi_0 | z | \psi_{n,l,m} \rangle \) is

\[
\langle \psi_{n',l',m'} | z | \psi_0 \rangle = \delta_{l',1} \delta_{m',0} \langle \psi_{n',1,0} | z | \psi_0 \rangle , \tag{A1}
\]

Second, the interaction Hamiltonian \( W \) (and hence, also the evolution operator which involves \( W \)) is a scalar of the rotation symmetry group. The matrix elements \( \langle \psi_{n,l,m} | W(s) | \psi_{n',l',m'} \rangle \) for \( l \neq l' \) and \( m \neq m' \) are then exactly equal to zero. Therefore, in the adiabatic approximation and due to the symmetry of the Hamiltonian \( \tilde{H} \), we obtain the relations

\[
\langle \psi_0 | T \exp \left( i \int_0^t ds W(-s) \right) | \psi_{n,l,m} \rangle \\
\approx \delta_{n,0} \delta_{l,0} \delta_{m,0} \langle \psi_0 | T \exp \left( -i \int_0^t ds W(s) \right) | \psi_0 \rangle , \tag{A2}
\]

\[
\langle \psi_{n',l',m'} | T \exp \left( -i \int_0^t ds W(s) \right) | \psi_{n',l',m'} \rangle \\
\approx \delta_{n'',n'} \delta_{l',l'} \delta_{m',m'} \langle \psi_{n',l',m'} | T \exp \left( -i \int_0^t ds W(s) \right) | \psi_{n',l',m'} \rangle . \tag{A3}
\]
Furthermore, because the ground state $\psi_0$ is non-degenerate, we find that
\[
\langle \psi_0 | T \exp \left( -i \int_0^t ds W(s) \right) | \psi_0 \rangle \approx 1,
\]
because within the adiabatic approximation, for any $n \geq 1$ the averages $\langle \psi_0 | W^n | \psi_0 \rangle = 0$.

The correlation function \[24\] using (A1) to (A3) takes the form
\[
f_{zz}(t) = \sum_n D_n e^{-i \Omega_{n,0} t} \times \langle \psi_{n,1,0} | 0_{ph} \langle T \exp \left[ -i \int_0^t ds W(s) \right] | 0_{ph} \rangle | \psi_{n,1,0} \rangle \tag{A4}
\]
with the squared matrix elements of the dipole transitions
\[
D_n \equiv |\langle \psi_{n,1,0} | z | \psi_0 \rangle|^2 = \frac{1}{3} \left( \int_0^\infty R_{n,1}(r) R_{0,0}(r) r^3 dr \right)^2, \tag{A5}
\]
and the FC transition frequencies
\[
\Omega_{n,0} \equiv E_{n,1} - E_0. \tag{A6}
\]
Further on, the interaction Hamiltonian is expressed in terms of the complex phonon coordinates $Q_k$:
\[
W = \sqrt{2} \sum_k W_k Q_k, \quad Q_k = \frac{b_k + b_k^+}{\sqrt{2}} \tag{A7}
\]
Here, we use the spherical-wave basis for phonon modes:
\[
\varphi_{k,l,m}(r) \equiv (-1)^{m-|m|/2} \phi_{k,l}(r) Y_{l,m}(\theta, \varphi), \tag{A8}
\]
where the radial part of the basis function is expressed through the spherical Bessel function $j_l(kr)$:
\[
\phi_{k,l}(r) = \left( \frac{2}{R} \right)^{1/2} k j_l(kr), \quad R = \left( \frac{3V}{4\pi} \right)^{1/3}. \tag{A9}
\]
The factor $(-1)^{m-|m|/2}$ is chosen in order to fulfil the symmetry property
\[
\varphi^*_{k,l,m}(r) = \varphi_{k,l,-m}(r).
\]
In the spherical-wave basis, the interaction Hamiltonian is
\[
W = \sqrt{2} \sum_{k,l,m} W_{k,l,m} Q_{k,l,m}, \tag{A10}
\]
with the complex phonon coordinates

\[ Q_{k,l,m} = \frac{b_{k,l,m} + b_{k,l,-m}^+}{\sqrt{2}} \]  (A11)

and with the interaction amplitudes

\[ W_{k,l,m} = \frac{\sqrt{2\sqrt{2}}}{k} (\varphi_{k,l,m}(r) - \rho_{k,l,m}) ; \rho_{k,l,m} \equiv \langle \psi_0 | \varphi_{k,l,m} | \psi_0 \rangle. \]  (A12)

The dipole-dipole correlation function (A4) is then

\[ f_{zz}(t) = \sum_n D_n e^{-i\Omega_{n,0}t} \]

\[ \times \left\langle \psi_{n,1,0} \left| 0_{ph} \right| \left( T \exp \left[ -i \int_0^t ds \sum_{k,l,m} W_{k,l,m}(s) Q_{k,l,m}(s) \right] \right| 0_{ph} \right| \psi_{n,1,0} \right. \]  (A13)

The operators \( W_{k,l,m}(s) \) in (A13) are equivalent to the \((2l + 1)\)-dimensional matrices \( \mathbb{W}_{k,l,m}^{(n)} \) determined in the basis of the level \((n, l)\). The matrix elements of these matrices are

\[ \left( \mathbb{W}_{k,l,m}^{(n)} \right)_{m_1,m_2}^{m_1,m_2} = \langle \psi_{n,1,m_1} | W_{k,l,m} | \psi_{n,1,m_2} \rangle. \]  (A14)

In these notations, \( f_{zz}(t) \) given by (A13) can be written down as

\[ f_{zz}(t) = \sum_n D_n e^{-i\Omega_{n,0}t} \left\langle \psi_{n,1,0} \left| 0_{ph} \right| \left( T \exp \left[ -i \int_0^t ds \mathbb{W}_{k,l,m}^{(n)}(s) \right] \right| 0_{ph} \right| 0_{ph} \right. \]  (A15)

where \( \mathbb{W}^{(n)} \) is the matrix electron-phonon interaction Hamiltonian expressed through the phonon complex coordinates in the spherical-wave representation as follows:

\[ \mathbb{W}^{(n)} = \sqrt{2} \sum_{k,l,m} \mathbb{W}_{k,l,m}^{(n)} Q_{k,l,m}. \]  (A16)

Here, \( \mathbb{W}_{k,l,m}^{(n)} \) is a \((3 \times 3)\) matrix in a basis of a level \((n, l)\) of the Hamiltonian \( \tilde{H}_0 \).

Because \( \mathbb{W}^{(n)} \) is a scalar of the rotation group, we can replace the diagonal matrix element of the T-exponent in (A15) with the trace in the aforesaid-finite-dimensional basis. As a result, we obtain for the polaron optical conductivity (4) with (A15) the expression

\[ \text{Re} \sigma(\omega) = \frac{\omega}{6} \sum_n D_n \int_0^\infty e^{i(\omega-\Omega_{n,0})t} \times \left\langle 0_{ph} \left| \text{Tr} \left( T \exp \left[ -i \int_0^t ds\mathbb{W}^{(n)}(s) \right] \right) \right| 0_{ph} \right| dt. \]  (A17)
Appendix B: Effective phonon modes

In order to perform the averaging in Eq. (30) analytically, we introduce the effective phonon modes $Q_{0,0}$ and $Q_{2,m}$ similarly to Ref. [26]. The Hamiltonian $\mathcal{W}^{(n)}$ in terms of these effective phonon modes is expressed as

$$\mathcal{W}^{(n)} = \sqrt{2} \sum_{l,m} \tilde{\mathcal{W}}_{l,m}^{(n)} Q_{l,m}$$

(B1)

where the matrices $\tilde{\mathcal{W}}_{l,m}^{(n)}$ (depending on the vibration coordinates $Q_{l,m}$) are explicitly given by the expressions (cf. Ref. [26]),

$$\mathcal{W}^{(n)} = a_0^{(n)} \mathbb{I} Q_{0,0} + a_2^{(n)} \sum_{m=-2}^{2} \mathbb{B}_m Q_{2,m}$$

(B2)

with the matrices $\mathbb{B}_j$

$$\mathbb{B}_0 = \frac{1}{2\sqrt{5\pi}} \begin{pmatrix} -1 & 0 & 0 \\ 0 & 2 & 0 \\ 0 & 0 & -1 \end{pmatrix},$$

(B3)

$$\mathbb{B}_1 = \mathbb{B}_1^+ = \frac{1}{2} \sqrt{\frac{3}{5\pi}} \begin{pmatrix} 0 & 0 & 0 \\ -1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix},$$

(B4)

$$\mathbb{B}_2 = \mathbb{B}_2^+ = \sqrt{\frac{3}{10\pi}} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}.$$}

(B5)

The coefficients $a_0^{(n)}$ and $a_2^{(n)}$ in Eq. (B2) are

$$a_0^{(n)} = \sqrt{2\alpha} \sum_{k} \frac{1}{k^2} \left[ \langle \phi_{k,0} \rangle_{n,1} - \langle \phi_{k,0} \rangle_{0,0} \right]^2 \right]^{1/2},$$

(B6)

$$a_2^{(n)} = \left( 4\sqrt{2\pi} \alpha \sum_{k} \frac{1}{k^2} \langle \phi_{k,2} \rangle_{n,1}^2 \right)^{1/2}.$$}

(B7)

Here $\phi_{k,l}$ is the radial part of the basis function expressed through the spherical Bessel function $j_l(kr)$:

$$\phi_{k,l}(r) = \left( \frac{2}{R} \right)^{1/2} k^l j_l(kr), \quad R = \left( \frac{3V}{4\pi} \right)^{1/3},$$

(B8)
V is the volume of the crystal, and $\langle f(r) \rangle_{n,l}$ is the average

$$
\langle f(r) \rangle_{n,l} = \int_0^\infty f(r) R_{n,l}^2(r) r^2 dr.
$$  \hspace{1cm} (B9)

The normalization of the phonon wave functions corresponds to the condition

$$
\int_0^R \phi_{k,l}(r) \phi_{k',l}(r) r^2 dr = \delta_{k,k'}.
$$  \hspace{1cm} (B10)

After the straightforward calculation using (B10), we express the coefficients $a_0^{(n)}$ and $a_2^{(n)}$ through the integrals with the radial wave functions:

$$
a_0^{(n)} = \left( 2 \sqrt{2} \alpha \int_0^\infty dr \int_0^r r (r')^2 \left[ R_{n,1}^2(r) - R_{0,0}^2(r) \right] \left[ R_{n,1}^2(r') - R_{0,0}^2(r') \right] \right)^{1/2},
$$  \hspace{1cm} (B11)

$$
a_2^{(n)} = \left( \frac{8 \sqrt{2} \pi \alpha}{5} \int_0^\infty dr \int_0^r (r')^4 r R_{n,1}^2(r) R_{n,1}^2(r') \right)^{1/2}.
$$  \hspace{1cm} (B12)

1. **Exact averaging**

Let us substitute the matrix interaction Hamiltonian (B2) to the dipole-dipole correlation function (30), what gives us the result

$$
f_{zz}(t) = \frac{1}{3} \sum_n D_n e^{-i \Omega_n t} \left\langle 0_{ph} \right| \exp \left( -it a_0^{(n)} Q_0 \right) \text{Tr} \exp \left( -it \frac{a_2^{(n)}}{2 \sqrt{5} \pi} V(Q_2) \right) \left| 0_{ph} \right\rangle.
$$  \hspace{1cm} (B13)

Here, we use the matrix depending on the phonon coordinates,

$$
V(Q_2) \equiv 2 \sqrt{5} \pi \sum_{m=-2}^2 B_m Q_{2m},
$$  \hspace{1cm} (B14)

whose explicit form is

$$
V(Q_2) = \begin{pmatrix}
-Q_{2,0} & -\sqrt{3} Q_{2,-1} & -\sqrt{6} Q_{2,-2} \\
-\sqrt{3} Q_{2,1} & 2 Q_{2,0} & \sqrt{3} Q_{2,-1} \\
-\sqrt{6} Q_{2,2} & \sqrt{3} Q_{2,1} & -Q_{2,0}
\end{pmatrix}.
$$  \hspace{1cm} (B15)

The matrix $V(Q_2)$ is analytically diagonalized. The equation for the eigenvectors $|\chi(Q_2)\rangle$ and eigenvalues $\lambda(Q_2)$ of $V(Q_2)$ is

$$
V(Q_2) |\chi(Q_2)\rangle = \lambda(Q_2) |\chi(Q_2)\rangle.
$$  \hspace{1cm} (B16)
The eigenvalues are found from the equation

\[ \det \left( V (Q_2) - \lambda (Q_2) I \right) = 0. \tag{B17} \]

We make the transformation to the real phonon coordinates,

\[
Q_{2,0} \equiv x_0, \\
Q_{2,m} \equiv \frac{x_m + iy_m}{\sqrt{2}}, \quad Q_{2,-m} = Q_{2,m}^* = \frac{x_m - iy_m}{\sqrt{2}}.
\]

Five variables \(x_0, x_1, x_2, y_1, y_2\) are the independent real phonon coordinates. The l.h.s. of Eq. (B17) is expressed in terms of these coordinates as

\[ \det \left( V (Q_2) - \lambda (Q_2) I \right) = -\lambda^3 + 3p\lambda + 2q \tag{B18} \]

with the coefficients

\[
p = x_0^2 + x_1^2 + x_2^2 + y_1^2 + y_2^2, \\
q = x_0^3 + \frac{3}{2} x_0 (x_1^2 + y_1^2) + \frac{3\sqrt{3}}{2} x_2 (x_1^2 - y_1^2) - 3x_0 (x_2^2 + y_2^2) + 3\sqrt{3}x_1y_1y_2.
\]

So, we have the cubic equation for \(\lambda\):

\[ \lambda^3 - 3p\lambda - 2q = 0. \tag{B19} \]

Because the matrix \(V (Q_2)\) is Hermitian, all its eigenvalues are real. Therefore, \(\frac{|q|}{p^{3/2}} \leq 1\) (otherwise, \(\sin (3\varphi)\) is not real). Herefrom, we have three explicit eigenvalues:

\[
\lambda_1 (Q_2) = 2\sqrt{p} \sin \left[ \frac{\pi}{3} + \frac{1}{3} \arcsin \left( \frac{q}{p^{3/2}} \right) \right], \\
\lambda_2 (Q_2) = -2\sqrt{p} \sin \left[ \frac{1}{3} \arcsin \left( \frac{q}{p^{3/2}} \right) \right], \\
\lambda_3 (Q_2) = -2\sqrt{p} \sin \left[ \frac{\pi}{3} - \frac{1}{3} \arcsin \left( \frac{q}{p^{3/2}} \right) \right]. \tag{B20}
\]

The trace in (B13) is invariant with respect to the choice of the basis. Consequently, after the diagonalization \(f_{zz} (t)\) takes the form

\[
f_{zz} (t) = \frac{1}{3} \sum_n D_n e^{-i \Omega_n t} \sum_{j=1}^{3} \left\langle 0_{ph} \left| \exp \left( -it \left[ a_0^{(n)} Q_0 + \frac{a_2^{(n)}}{2\sqrt{5\pi}} \lambda_j (Q_2) \right] \right) \right| 0_{ph} \right\rangle. \tag{B21}
\]
After inserting \( f_z(t) \) given by (B21) into (4), the integration over time gives the delta function multiplied by \( 2\pi \), and we arrive at the result
\[
\text{Re} \sigma(\omega) = \frac{\pi \omega}{3} \sum_n D_n \sum_{j=1}^3 \left\langle 0_{ph} \left| \delta \left( \omega - \Omega_{n,0} - a_0^{(n)} Q_0 - \frac{a_0^{(n)}}{2\sqrt{5\pi}} \lambda_j (Q_2) \right) \right| 0_{ph} \right\rangle. \quad (B22)
\]
The ground-state wave function for the effective phonon modes is
\[
|0_{ph}\rangle \equiv \Phi_0(Q) = \Phi_0^{(0)}(0) \Phi_0^{(2)}(Q_2). \quad (B23)
\]
\( \Phi_0^{(0)}(0) \) is the one-oscillator ground-state wave function:
\[
\Phi_0^{(0)}(0) = \frac{\pi}{4} \exp \left( -\frac{Q_0^2}{4} \right). \quad (B24)
\]
The ground-state wave function of phonons with \( l = 2 \) is:
\[
\Phi_0^{(2)}(Q_2) = \frac{\pi}{4} \exp \left[ -\frac{1}{2} \left( x_0^2 + \sum_{m=1,2} (x_m^2 + y_m^2) \right) \right]. \quad (B25)
\]
The phonon ground-state wave function (B23) is then
\[
\Phi_0(Q) = \frac{1}{\pi^{3/2}} \exp \left[ -\frac{1}{2} \left( x_0^2 + \sum_{m=1,2} (x_m^2 + y_m^2) + Q_0^2 \right) \right]. \quad (B26)
\]
With these phonon wave functions, Eq. (B22) results in the following expression for the polaron optical conductivity
\[
\text{Re} \sigma(\omega) = \frac{\omega}{3\pi^2} \sum_n D_n \left\langle 0_{ph} \left| \int_{-\infty}^{\infty} dx_0 \int_{-\infty}^{\infty} dx_1 \int_{-\infty}^{\infty} dx_2 \int_{-\infty}^{\infty} dy_1 \int_{-\infty}^{\infty} dy_2 \left\langle 0_{ph} \left| \delta \left( \omega - \Omega_{n,0} - a_0^{(n)} Q_0 - \frac{a_0^{(n)}}{2\sqrt{5\pi}} \lambda_j (Q_2) \right) \right| 0_{ph} \right\rangle \right. \right. \nonumber
\]
\[
\times \sum_{j=1}^3 \left[ x_0^2 + \sum_{m=1,2} (x_m^2 + y_m^2) + \frac{\left( \omega - \Omega_{n,0} - \frac{a_0^{(n)}}{2\sqrt{5\pi}} \lambda_j (Q_2) \right)^2}{\left( a_0^{(n)} \right)^2} \right] \right\rangle. \quad (B27)
\]

2. Averaging neglecting the Jahn-Teller effect

In order to perform the phonon averaging explicitly, we disentangle the exponent
\[
\exp \left( -i \sqrt{2} \sum_{l,m} \bar{W}^{(n)}_{l,m} Q_{l,m} \right)
\]
as follows.
\[
\exp \left( -i \sqrt{2} \sum_{l,m} \bar{W}^{(n)}_{l,m} Q_{l,m} \right) = \exp \left( -i \sum_{l,m} \bar{W}^{(n)}_{l,m} b^+_l b_m \right)
\]
\[
\times \text{T} \exp \left( -i \int_0^t ds \sum_{l,m} e^{i s \sum_{l',m'} \bar{W}^{(n)}_{l',m'} b^+_{l',m'} b_{l,m} e^{-i s \sum_{l',m'} \bar{W}^{(n)}_{l',m'} b^+_l b_{l',m'}}} \right). \quad (B28)
\]
Neglecting non-commutation of matrices $\tilde{W}_{l,m}^{(n)}$ we find that

$$
\sum_{l,m} e^{is \Sigma_{l',m'} \tilde{W}_{l',m'}^{(n)} b_{l',m'}^+ \tilde{W}_{l,m}^{(n)} b_{l,m}} e^{-is \Sigma_{l',m'} \tilde{W}_{l',m'}^{(n)} b_{l',m'}^+ b_{l,m}} = \sum_{l,m} \tilde{W}_{l,m}^{(n)} b_{l,m} - is \sum_{l,m} \tilde{W}_{l,-m}^{(n)} \tilde{W}_{l,m}^{(n)}. \tag{B29}
$$

The sum $\sum_{l,m} \tilde{W}_{l,-m}^{(n)} \tilde{W}_{l,m}^{(n)}$ in the basis $(l,m)$ for a definite $n$ is proportional to the unity matrix. Therefore,

$$
\exp\left(-it \sqrt{2} \sum_{l,m} \tilde{W}_{l,m}^{(n)} Q_{l,m}\right) \text{ is } e^{-it \sqrt{2} \sum_{l,m} \tilde{W}_{l,m}^{(n)} Q_{l,m}}
$$

that gives us the result

$$
\langle 0_{ph} \mid e^{-it \sqrt{2} \sum_{l,m} \tilde{W}_{l,m}^{(n)} Q_{l,m}} \mid 0_{ph} \rangle = e^{-\frac{t^2}{2} \Sigma_{l,m} \tilde{W}_{l,-m}^{(n)} \tilde{W}_{l,m}^{(n)}.} \tag{B30}
$$

Using the explicit formulae for the matrices $\tilde{W}_{l,m}^{(n)}$, the matrix sum takes the form

$$
\sum_{l,m} \tilde{W}_{l,-m}^{(n)} \tilde{W}_{l,m}^{(n)} = \omega_s^{(n)} \mathbb{I} \tag{B32}
$$

with the parameter

$$
\omega_s^{(n)} = \frac{1}{2} \left( a_0^{(n)} \right)^2 + \frac{1}{4\pi} \left( a_2^{(n)} \right)^2. \tag{B33}
$$

Using (B32), the optical conductivity [30] is transformed to the expression

$$
\text{Re} \sigma(\omega) = \omega \sum_n \sqrt{\frac{\pi}{2S_n}} D_n \exp\left(-\frac{(\omega - \Omega_{n,0})^2}{2S_n}\right). \tag{B34}
$$

[1] J. T. Devreese and A. S. Alexandrov, Rep. Prog. Phys. 72, 066501 (2009).
[2] V. Gurevich, I. Lang, and Yu. Firsov, Sov. Phys. Solid State 4, 918 (1962).
[3] J. Devreese, W. Huybrechts, and L. Lemmens, Phys. Stat. Sol. (b) 48, 77 (1971).
[4] W. Huybrechts and J. T. Devreese, Phys. Rev. B 8, 5754 (1973).
[5] B. E. Sernelius, Phys. Rev. B 48, 7043 (1993).
[6] E. Kartheuser, R. Evrard, and J. Devreese Phys. Rev. Lett. 22, 94-97 (1969).
[7] M. J. Goovaerts, J. M. De Sitter, and J. T. Devreese, Phys. Rev. B 7, 2639 (1973).
[8] J. Devreese, J. De Sitter, and M. Goovaerts, Phys. Rev. B 5, 2367 (1972).
[9] R. Feynman, R. Hellwarth, C. Iddings, and P. Platzman, Phys. Rev. 127, 1004 (1962).
[10] R. P. Feynman, Phys. Rev. 97, 660 (1955).
[11] J. T. Devreese, in Polarons in Ionic Crystals and Polar Semiconductors (North-Holland, Amsterdam, 1972), pp. 83 – 159.
[12] F. M. Peeters and J. T. Devreese, Phys. Rev. B 28, 6051 (1983).
[13] J. Devreese and R. Evrard, Phys. Lett. 11, 278 (1964).
[14] A. S. Mishchenko, N. V. Prokof’ev, A. Sakamoto, and B. V. Svistunov, Phys. Rev. B 62, 6317 (2000).
[15] A.S. Mishchenko, N. Nagaosa, N. V. Prokof’ev, A. Sakamoto, and B. V. Svistunov, Phys. Rev. Lett. 91, 236401 (2003).
[16] G. De Filippis, V. Cataudella, A. S. Mishchenko, C. A. Perroni, and J. T. Devreese, Phys. Rev. Lett. 96, 136405 (2006).
[17] L. D. Landau and S. I. Pekar, J. Exp. Theor. Phys. 18, 419 (1948).
[18] G. R. Allcock, in Polarons and Excitons, edited by C. G. Kuper and G. D. Whitfield (Oliver and Boyd, Edinburgh, 1963), pp. 45 – 70.
[19] M. Born and K. Huang, Dynamical theory of crystal lattices (Oxford University Press, 2007).
[20] Yu. E. Perlin, Sov. Physics. Uspekhi. 6, 542 (1964).
[21] S. I. Pekar, Untersuchungen über die Elektronentheorie der Kristalle (Akademie-Verlag, Berlin, 1954).
[22] S. J. Miyake, J. Phys. Soc. Jap. 38, 181 (1975); 41, 747 (1976).
[23] H. Kleinert, Path Integrals in Quantum Mechanics, Statistics, Polymer Physics, and Financial Markets (5th edition, World Scientific, Singapore 2009).
[24] H. Jahn and E. Teller, Proc. R. Soc. London A 161, 220 (1937).
[25] J. T. Devreese, L. F. Lemmens, J. Van Royen, Phys. Rev. B 15, 1212 (1977).
[26] V. M. Fomin, V. N. Gladilin, J. T. Devreese, E. P. Pokatilov, S. N. Balaban, and S. N. Klimin, Phys. Rev. B 57, 2415 (1998).
[27] D. Emin, Phys. Rev. B 48, 13691 (1993).
[28] E. N. Myasnikov, A. E. Myasnikova, and Z. P. Mastropas, Physics of the Solid State 48, 1046 (2006).
[29] H. Spohn, Phys. Rev. B 33, 8906 (1986); Ann. Phys. 175, 278 (1987).
Figure captions

Fig. 1. Frequency of the main peak in the optical conductivity spectra calculated within the model of Ref. [8] (dots) and the main-peak energy extracted from the DQMC data [15, 16] (squares).

Fig. 2. The strong-coupling polaron optical conductivity calculated within the rigorous strong-coupling approach of the present work (solid curves), within the present approach but neglecting the Jahn – Teller effect (dashed curves), within the adiabatic approximation of Ref. [16] (dot-dashed curves), and the numerical Diagrammatic Monte Carlo data (full dots) for $\alpha = 8$ and 8.5. The FC and RES transition frequencies are indicated by the arrows.

Fig. 3. The strong-coupling polaron optical conductivity calculated within the rigorous strong-coupling approach of the present work (solid curves), within the present approach but neglecting the Jahn – Teller effect (dashed curves), within the adiabatic approximation of Ref. [16] (dot-dashed curves), and the numerical Diagrammatic Monte Carlo data (full dots) for $\alpha = 9, 13$ and 15. The FC and RES transition frequencies are indicated by the arrows.
Fig. 1  S. N. Klimin, J. T. Devreese
Fig. 2  S. N. Klimin, J. T. Devreese
Fig. 3  S. N. Klimin, J. T. Devreese