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Optical conductivity of the Fröhlich polaron

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We present accurate results for optical conductivity of the three dimensional Fröhlich polaron in all coupling regimes. The systematic-error free diagrammatic quantum Monte Carlo method is employed where the Feynman graphs for the momentum-momentum correlation function in imaginary time are summed up. The real-frequency optical conductivity is obtained by the analytic continuation with stochastic optimization. We compare numerical data with available perturbative and non-perturbative approaches to the optical conductivity and show that the picture of sharp resonances due to relaxed excited states in the strong coupling regime is “washed out” by large broadening of these states. As a result, the spectrum contains only a single-maximum broad peak with peculiar shape and a shoulder.

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Since the seminal work of Landau [1] the polaron problem, i.e. renormalization of quasiparticle properties due to coupling to phonons, has been attracting considerable attention serving as a testing ground for non-perturbative methods. The Hamiltonian for polarons consists of terms describing free particle and phonon states [2] (Planck's constant and electric charge are set to unity)

$$H_0 = \sum_{\mathbf{k}} \varepsilon(\mathbf{k}) a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \sum_{\mathbf{q}} \omega_{\mathbf{q}} b_{\mathbf{q}}^\dagger b_{\mathbf{q}} \quad (1)$$

which interact through the particle-phonon coupling

$$H_{\text{e-ph}} = \sum_{\mathbf{k}, \mathbf{q}} \left(V(\mathbf{q}) b_{\mathbf{q}}^\dagger a_{\mathbf{k}-\mathbf{q}}^\dagger a_{\mathbf{k}} + H.c. \right) . \quad (2)$$

In Eqs. (1-2), $a_{\mathbf{k}}$ and $b_{\mathbf{q}}$ are the particle and phonon annihilation operators in momentum space, respectively. The most popular models are the following two: (i) Holstein lattice polaron (characterized by the tight-binding dispersion law $\varepsilon(\mathbf{k}) = 2t \sum_{i=x,y,z} \cos(k_i)$ with hopping amplitude t , dispersionless optical phonons $\omega_{\mathbf{q}} = \omega_0$, and short-range interaction vertex $V_{\text{H}}(\mathbf{q}) = \gamma$), and (ii) the continuous Fröhlich polaron (FP) (characterized by $\varepsilon(\mathbf{k}) = k^2/2m$, $\omega_{\mathbf{q}} = \omega_0$, and long-range interaction $V_{\text{F}}(\mathbf{q}) = i\omega_0 [2\sqrt{2}\pi\alpha\sqrt{m\omega_0}]^{1/2}/q$; the convention is to measure energies in units of ω_0 , and length scales in units of $1/\sqrt{m\omega_0}$).

Although there are many reliable methods of calculating ground-state properties of polarons, the latter are hard to test directly in experiments. In contrast, the experimental data on optical conductivity (OC) spectra, $\sigma_{\beta\delta}(\omega)$, are readily available, but most theoretical methods can not well treat the excited states, and, as a result, are less accurate in predicting $\sigma(\omega)$. For decades, going back to early papers [2, 3, 4], the polaron OC (especially within the Fröhlich model [5, 6, 7, 8]) constantly attracts attention of theoretical community.

Even in the strong-coupling limit the results on OC are full of contradictions. In the variational adiabatic treatment of polarons at strong coupling [9] one finds the so-called relaxed excited states (RES), i.e. quasi-stable states with the lattice distortion adjusted to the excited particle wavefunction. To contribute sharp peaks to OC spectra the decay rate of the lowest RES must be relatively small (otherwise the very notion of RES becomes vague). The decay rate for RES with the energy $0.066\alpha^2$ [10, 11, 12] was calculated in the one-phonon approximation [6] and a sharp peak in OC was predicted for $\alpha \geq 5$. Devreese, De Sitter, and Goovaerts (DSG) [7] reproduced this result by the expansion of the impedance $Z(\omega) \sim 1/\sigma(\omega)$ within the Feynman-Hellwarth-Iddings-Platzman (FHIP) theory [3]. However, the validity of the one-phonon approximation [6] is in doubt for the strong coupling limit. Also, even within the *same FHIP approximation* [3], the expansion of the inverse impedance does not show sharp features at energies predicted by the impedance expansion [7]. Finally, recent high-precision simulations of the Lehman spectral function [13], $g(\omega)$, did not reveal any stable excited states for FP with the α^2 -dependence of energy - peaks with weak energy dependence on α were found instead. It is worth mentioning, that the perturbation theory fails to reproduce data on $g(\omega)$ even at very small $\alpha = 0.05$.

At present, the actual shape of OC for the Fröhlich polaron is not known, and the crucial link connecting polaron theories to experiments is missing. We are not aware of any systematic-error free method for calculating OC of continuous polarons. There are several non-perturbative approaches [13, 14, 15] dealing with the properties determined by excited states. However, the Lanczos diagonalization technique [15] is limited to finite size systems, and the variational method [14] encounters difficulties for long-range interactions.

In this Letter, we show that the problem may be solved by the Diagrammatic Monte Carlo method [13] based on

direct summation of Feynman diagrams for the particle momentum-momentum correlation function in imaginary time, $\langle k_\beta(\tau)k_\delta(0) \rangle$, with subsequent analytic continuation to real frequencies by means of stochastic optimization. Our technique is generic and works for arbitrary particle and phonon dispersion laws and interaction vertex. For the FP model we find that existing analytical methods severely underestimate multiphonon decay rates which broaden RES to the point that their contributions to OC overlap. Instead of sharp peaks one obtains a picture of a broad single-maximum function with irregular shape and a shoulder.

The real part of $\sigma_{\beta\delta}(\omega)$ is straightforwardly related to $\langle k_\beta(\tau)k_\delta(0) \rangle$

$$\sigma_{\beta\delta}(\omega) = (\pi/\omega)\langle k_\beta k_\delta \rangle_\omega, \quad (3)$$

where (at zero temperature $\beta = 1/T \rightarrow \infty$)

$$\langle k_\beta(\tau)k_\delta(0) \rangle = \int_0^{+\infty} d\omega e^{-\omega\tau} \langle k_\beta k_\delta \rangle_\omega. \quad (4)$$

The solution of the integral Eq. (4) is obtained by the stochastic optimization method of Ref. [13, 16] which allows to handle both broad and infinitely sharp features in the spectrum on equal footing. The $\langle k_\beta(\tau)k_\delta(0) \rangle$ correlator is diagonal in the particle momentum representation and is readily available (has a direct Monte Carlo estimator) from the statistics of Feynman diagrams for the partition function, $Z = Tr \{ e^{-\beta H} \} \equiv Tr \{ e^{-\beta H_0} \hat{T}_\tau \exp [- \int_0^\beta H_{e-ph}(\tau) d\tau] \}$, (see Fig. 1).

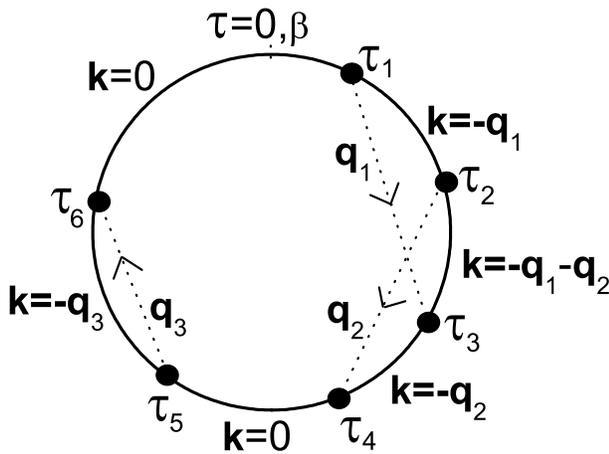


FIG. 1: A typical diagram for the polaron partition function. Solid (dotted) lines represent free particle (phonon) propagators. In general, there will be diagrams with all particle propagators being accompanied by one or several phonon lines.

Each graph generated by the expansion of the τ -ordered exponent contributes $\beta^{-1} \int_0^\beta d\tau' k_\beta(\tau' + \tau)k_\delta(\tau')$ to the correlator, where $\mathbf{k}(\tau')$ is the step-wise function on the diagram circle. All calculations were done for

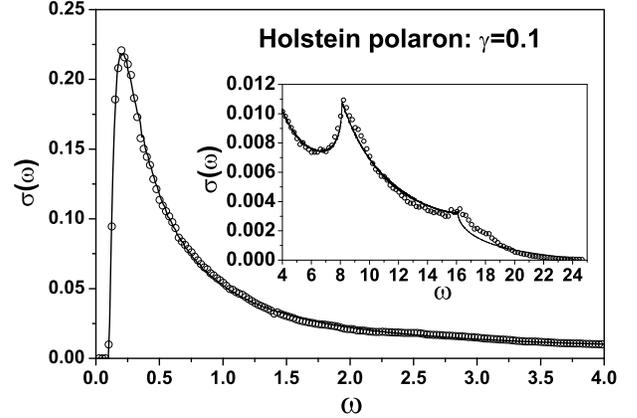


FIG. 2: The low-energy part of OC for the Holstein polaron (circles) compared to the perturbation theory result (line); the high-energy tail is shown in the insert.

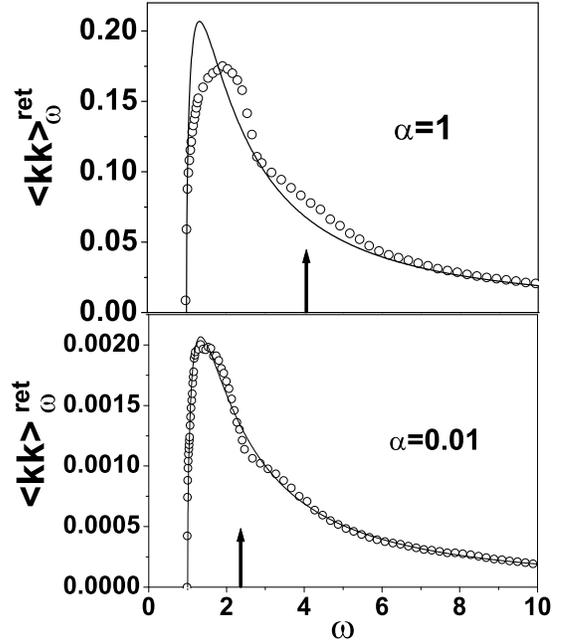


FIG. 3: Comparison between the calculated momentum-momentum correlator (circles) and the perturbation theory result (lines) for the Fröhlich model. We indicate by arrows anomalies observed previously in the Lehman function.

finite but very large $\beta\omega > 100$, to ensure that exponentially small finite-temperature corrections are negligible. The Metropolis-type sampling of diagrams in continuous momentum-time was performed using Diagrammatic Monte Carlo technique developed in Refs. [13, 17]. We note, that formally our method works for finite temperatures as well, but analytic continuation is notoriously unstable at high temperatures ($\omega_0 T \sim 1$ in our case).

To test the method efficiency and reliability for

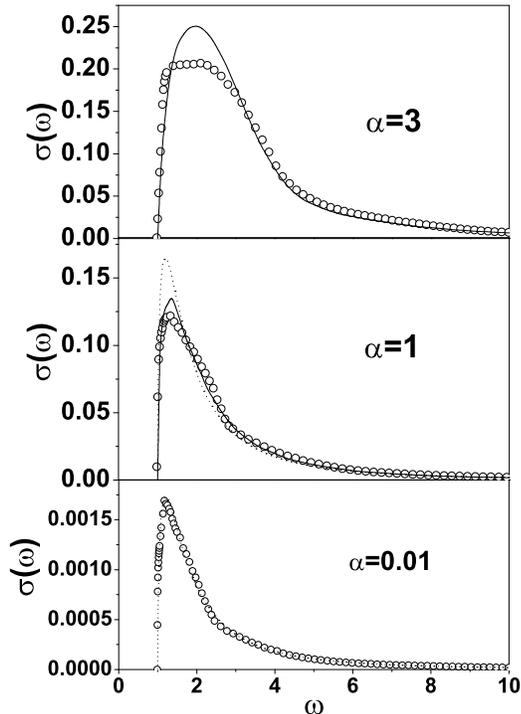


FIG. 4: OC data for the weak coupling regime (open circles) compared to the second-order perturbation theory (dotted lines) and DSG calculations (solid lines).

the study of OC, we simulated the three dimensional Holstein polaron in the weak coupling regime ($t = 2$, $\omega_0 = 0.1$, $\gamma = 0.1$) and compared our data in Fig. 2 to the perturbation theory result [18], $\sigma_{\alpha\alpha}(\omega) = \sigma(\omega) = (\gamma^2/8\pi^2\omega) \int_{\text{BZ}} d^3q q_\alpha^2 \delta(\omega - \omega_0 - \varepsilon(\mathbf{q}))$, where the integral is taken over the Brillouin zone. Not only the low-energy part is in perfect agreement with the theory, but even sharp features in the tail due to van Hove singularities are reproduced with remarkable (for analytic continuation procedures) accuracy.

There is no such agreement with the perturbation theory for the Fröhlich polaron even at extremely small coupling constant $\alpha = 0.01$ (see Fig. 3). This is due to the singular $1/q$ -dependence of the coupling constant. We note that positions of anomalies in $\sigma(\omega)$ and $g(\omega)$ (see Ref. [13]) spectra correlate. Apart from weak anomaly around $\omega \sim 3$, the OC spectrum is in reasonable overall agreement with the second-order perturbation theory result for $\alpha = 0.01$ (see Fig. 4).

For $\alpha = 1$ our results deviate from the 2-nd order perturbation theory considerably; the agreement with the DSG method of impedance expansion [7] is much better. Since fourth-order corrections are very small in this parameter range and do not lead to the DSG curve [10], we conclude that two-phonon terms are not sufficient to account for the discrepancy and higher-order terms have to be taken into account even at $\alpha = 1$. For $\alpha = 3$ the spectrum broadens considerably and its shape in the

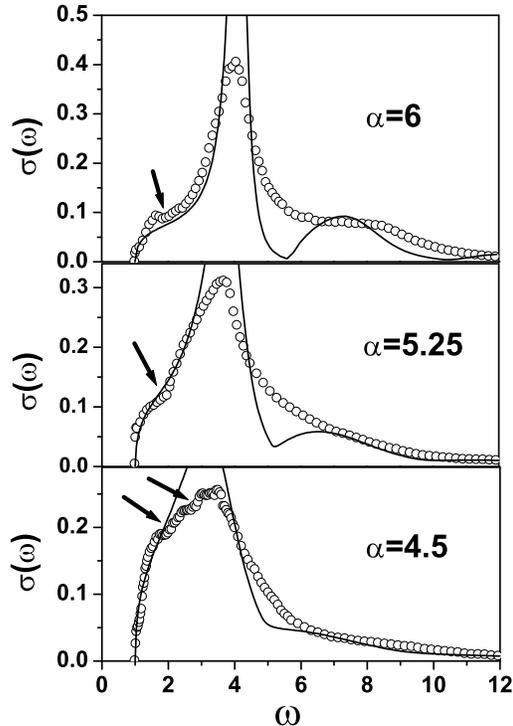


FIG. 5: OC data for the intermediate coupling regime (open circles) compared to the DSG approach (solid line). Arrows point to the anomalies in absorption spectra arising at the two- and three-phonon thresholds.

maximum starts deviating from the DSG result.

In the intermediate coupling regime $3 < \alpha < 7$ we observe that the position of the spectrum maximum does follow the DSG prediction (and that of other papers advocating the RES concept [6, 10, 11, 12]) for the largest peak, and the low-energy behavior is also captured by the theory reasonably well. However, the second peak at higher frequencies never actually fully develops, and, at best, its remnants are seen as a flat shoulder for $\alpha = 6$. The other qualitative difference between our results and DSG theory is significantly larger broadening of the dominant peak. We stress, that broadening in our data is not an artifact of the analytic continuation method because we clearly see thresholds corresponding to two- and three-phonon emission/absorption processes (see Fig. 5). The conclusion we draw from this observation is that excited states are short-lived and their decay rates mediated by multi-phonon processes are not accounted for in [6, 10, 11, 12].

In the strong coupling regime, the broadening of excited states completely changes the overall shape of the spectrum (see Fig. 6). Instead of the double-peak structure with extremely narrow central peak and a pseudogap, $\sigma(\omega)$ features a broad single-maximum function away from the threshold. Even more surprisingly, the

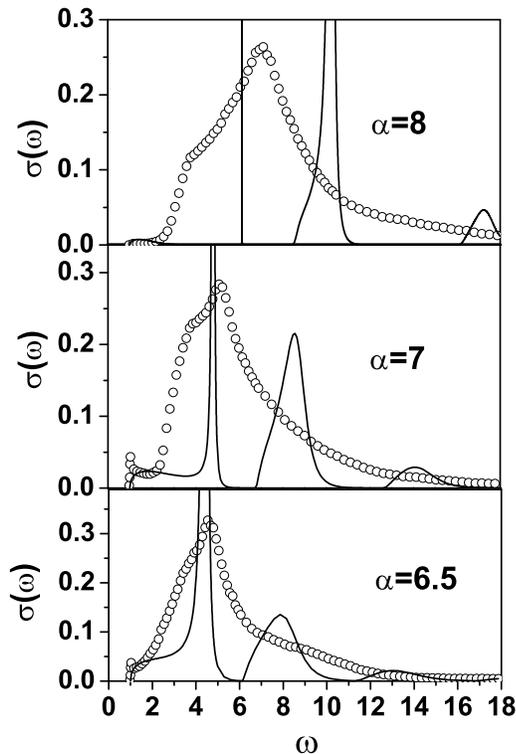


FIG. 6: OC data for the strong coupling regime (open circles) compared to the DSG results (solid lines).

shoulder at the high-frequency end of the spectrum vanishes, and another shoulder emerges at the low-frequency

end (but away from the threshold at $\omega = \omega_0$). The central peak and the new shoulder can be fitted (somewhat arbitrarily) as resulting from two strongly overlapping Gaussian peaks. In any event, the concept of quasi-stable excited states clearly has no place even at large $\alpha = 8$ (the effective mass is already heavily renormalized at this point by a factor of thirty [13]).

We conclude that physically attractive picture of stable RES, naturally emerging from the variational treatment of the strong coupling limit, may not be used to interpret OC spectra of Fröhlich polarons. Decay rates of excited states are severely underestimated in the one-phonon approximation [6, 10]. We may not rule out the possibility that at larger values of α sharp peaks do appear in OC. However, this outcome would be rather academic because for $\alpha > 15$ the effective mass renormalization is enormous, $m^*(\alpha = 15) \sim 10^3 m$, and lattice effects enter the problem. In realistic models of polarons one also has to include into the picture (i) several optical modes, (ii) interactions with acoustic phonons, (iii) screening of the interaction potential, (iv) finite lattice constant, etc. The simulation method developed in this Letter allows one to study all of the above mentioned effects. Therefore the accurate analysis of the experimental data is possible, which is left for future studies. We note, that formally our method works for finite temperatures as well, but analytic continuation is notoriously unstable at high temperatures ($\omega_0 T \sim 1$ in our case).

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