Quadratic electron-phonon coupling by Diagrammatic Monte Carlo

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Abstract

In recent years, a growing interest in anharmonic effects in materials arose due to the expected impact of anharmonic electron-phonon interactions on phonon-mediated superconductivity in hydrides and on polaron mobility in quantum paraelectrics [1] and halide perovskites [2]. This has led to the development of model Hamiltonians [1], [3] which take into account the anharmonicity of the lattice and go beyond the usual linear electron-phonon coupling.

In this work, we consider a modified Holstein Hamiltonian with a quadratic electron-phonon interaction term [4]. The Hamiltonian is solved numerically by Diagrammatic Monte Carlo, calculating zero temperature GS properties such as polaron energy, average phonon number and effective mass. We benchmark our results with the atomic limit where analytic solutions are available. We conclude by discussing the extension to finite temperature effects and future directions.

Polaron model

We take the Holstein Hamiltonian (tight binding electron dispersion, one optical phonon branch) and replace the usual linear electron-phonon interaction with a term quadratic in the lattice displacement.

$$\widehat{H}_{\text{el}} = \sum_{\boldsymbol{k}} \epsilon(\boldsymbol{k}) a^\dagger_{\boldsymbol{k}} a_{\boldsymbol{k}} \quad \epsilon(\boldsymbol{k}) = 2t\sum_{i=1}^{d}(1-\cos(k_i a)) \quad -\pi/a \le k_i < \pi/a$$

$$\widehat{H}_{\text{ph}} = \sum_{\mathbf{q}} \omega(\mathbf{q}) b^\dagger_{\mathbf{q}} b_{\mathbf{q}} \qquad \omega(\mathbf{q}) = \Omega_0$$

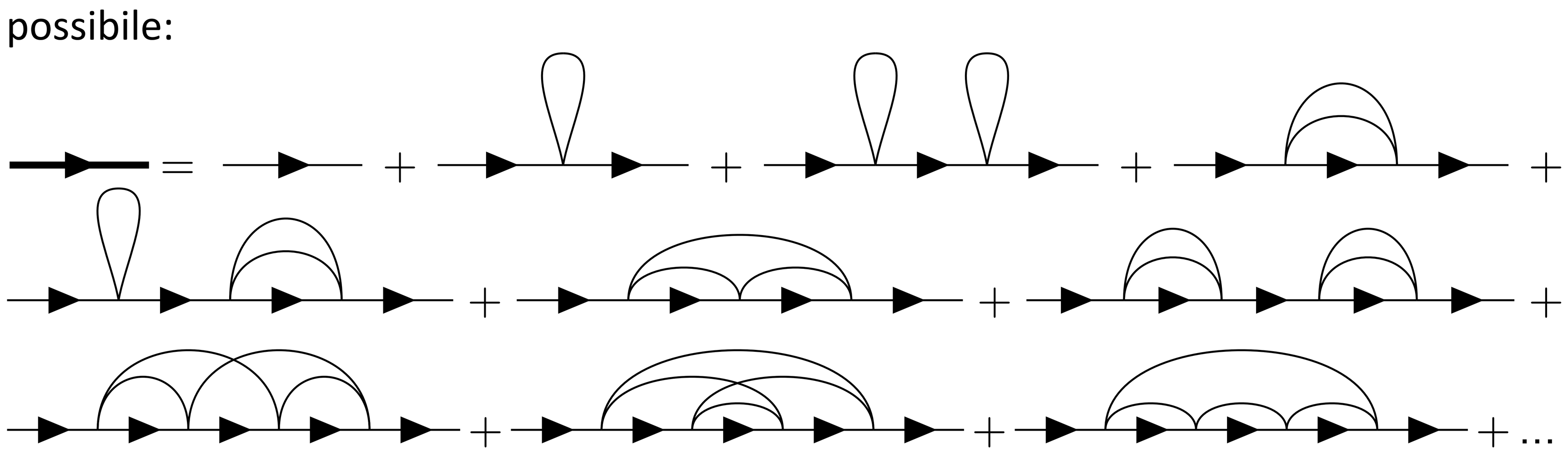
$$\widehat{H}_{\text{el-ph-2}} = \frac{1}{\sqrt{N^2}} \sum_{\mathbf{k},\mathbf{q}_1,\mathbf{q}_2} g_2\, a^\dagger_{\mathbf{k}+\mathbf{q}_1+\mathbf{q}_2} a_{\mathbf{k}} (b^\dagger_{-\mathbf{q}_1} + b_{\mathbf{q}_1})(b^\dagger_{-\mathbf{q}_2} + b_{\mathbf{q}_2})$$

If $\widehat{H}_{\text{el}}$ is omitted ($t=0$), we speak of the **atomic limit**, where an analytic solution is well known, providing an extremely useful benchmark for our data. A canonical transformation is applied to transform the position-space Hamiltonian into an effective harmonic oscillator with renormalized frequency Ω_{at} and shifted ground state energy [4].

$$\widehat{H}_{\text{at}} = \Omega_{\text{at}}\gamma^\dagger\gamma + \frac{\Omega_{\text{at}}-\Omega_0}{2} \quad \Omega_{\text{at}} = \sqrt{\Omega_0(\Omega_0+4g_2)} \quad Z_0 = \sqrt{\frac{\Omega_0+2g_2+\Omega_{\text{at}}}{2\Omega_{\text{at}}}}^{-1}$$

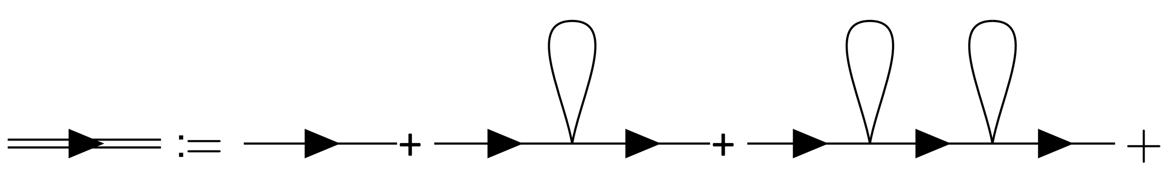
Methodology

The Hamiltonian is solved exactly by Diagrammatic Monte Carlo (DiagMC), a random sampling of Feynman diagrams which constitute the one-electron imaginary time Green function. Due to the extended interaction, a broader set of diagrams and topologies is possibile:



All the ground state properties can be extracted from the large imaginary time behaviour of the Green function.

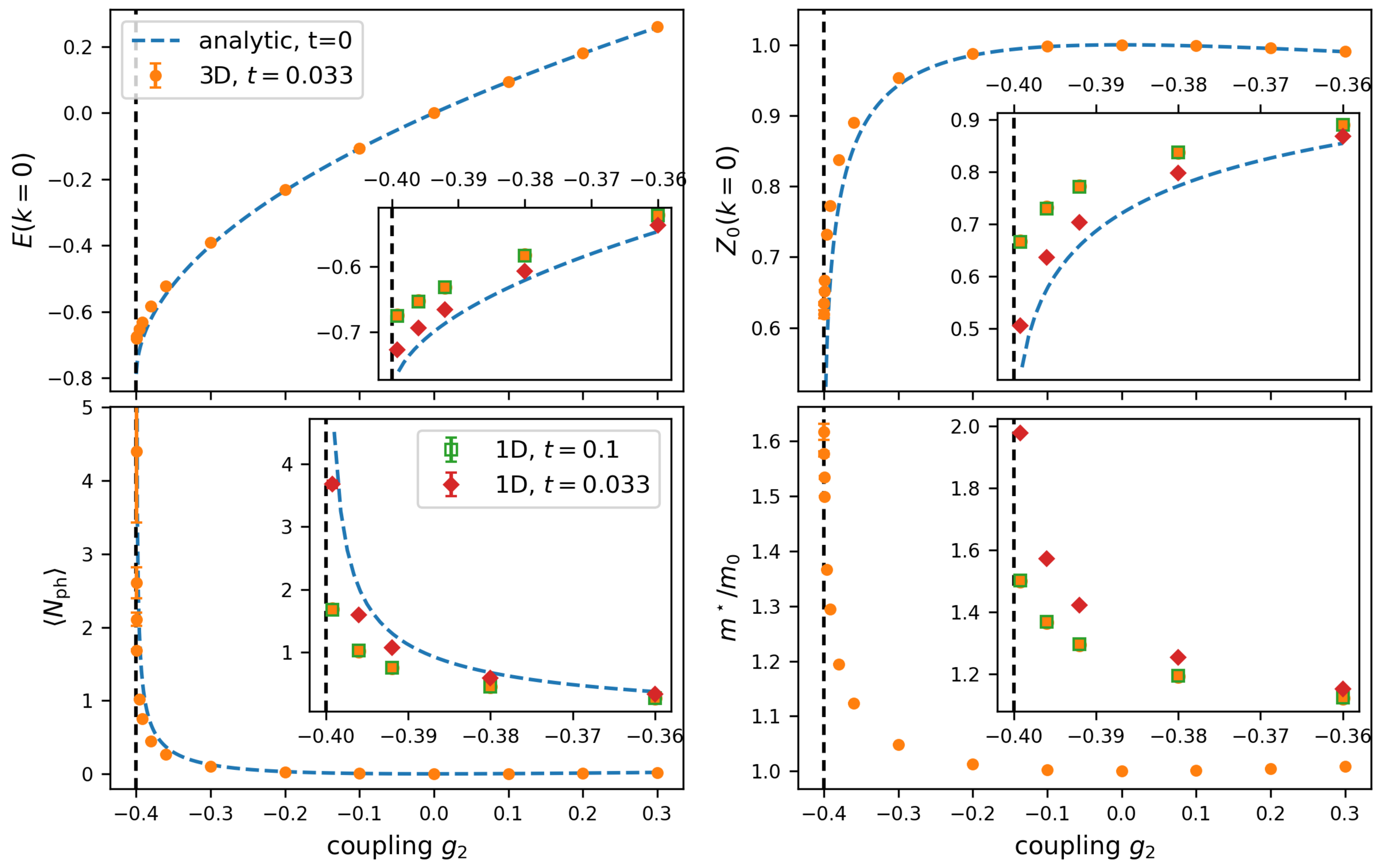
The sign of the diagram is given by $(-\text{sgn}(g_2))^n$, where n is the diagram order. It is always positive if $g_2 < 0$, but for $g_2 > 0$, it is negative whenever n is odd. This sign problem is mainly caused by single phonon loops. The series of the single phonon loop diagrams can be summed analytically and included in a renormalized electron propagator.



The same technique can also be applied to study finite temperature effects by employing the temperature phonon propagator. The total diagram length then acquires the interpretation of inverse temperature. In this case we measure the current-current correlation function in Matsubara frequency and obtain the mobility through numerical analytic continuation. The form of the current operator used is $\widehat{j} = 2eat\sum_k sin(ka)c_k c_k$, derived from the Peierls substitution for lattice models.

Ground state properties

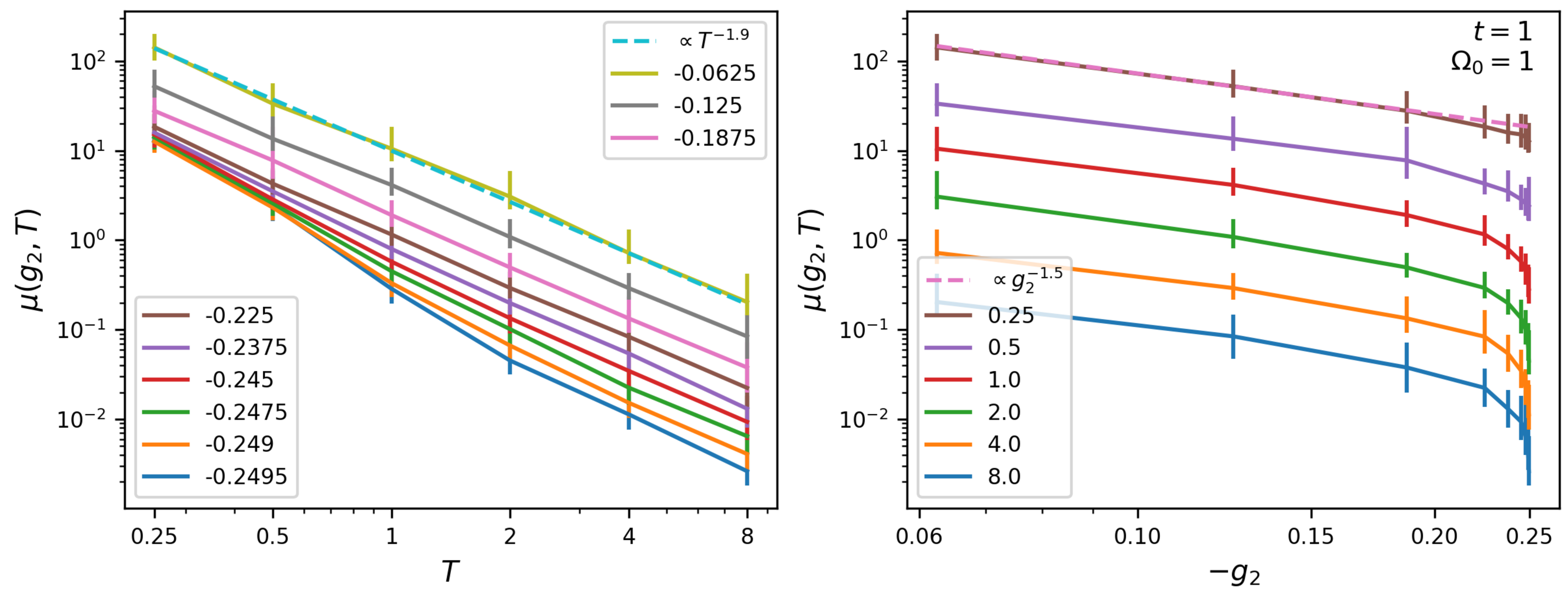
At zero T and $\Omega_0 = 1.6$, we probed the ground state by measuring the polaron binding energy, quasiparticle weight, average phonon number and effective mass for different hoppings and dimensions, benchmarking them with the known atomic limit results.



At positive coupling, the renormalization is very weak and close to the atomic limit. At negative coupling, the atomic limit divergence at $\Omega_0/4$ is retained also at small finite hopping. At each coupling, increasing the hopping decreases the renormalization, leading to a steeper divergency. The $3D$, $t=0.033$ points overlap with the $1D$, $t=0.1$ data due to having the same electron bandwidth. Compared to the linear interaction case, convergence is harder to achieve for the same number of phonons in the cloud.

Temperature and coupling effects on mobility

We studied the behavior of static mobility against temperature and coupling in the negative g_2 regime. We plot both axes in log scale to identify typical power-law dependencies.



The mobility was obtained from the Matsubara frequency current-current correlation function via Stochastic Optimization with Consistent Constraints, which is free from any artificial bias [5]. At low coupling a dependence $\mu \propto T^{-\delta}$ is found, with an exponent $\delta = 1.9$ close to the $\delta = 2$ value of the linear interaction case. Compared to the linear coupling [6], no regime where the derivative changes sign was observed.

Outlook and future work

The goal of the present study was to isolate the novel quadratic electron-phonon interaction and investigate its effects on the ground state and temperature dependent properties with DiagMC. This work represents the first step towards the solution of more complicated Hamiltonians that feature the quadratic interaction, such as the model proposed in [1] for STO, where electrons are coupled to a soft transverse optical mode via the two-phonon mechanism. The same form also appears in the extended anharmonic Fröhlich Hamiltonian derived in [3], which enables the connection to first-principle inputs.

References

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