Finite-temperature dynamical properties of the Holstein model: Hierarchical equations of motion approach

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Polaron problem: The oldest condensed-matter problem

- properties (static, **dynamic**) of an electron coupled to lattice vibrations (≈ 90 yrs.)

Holstein model–bootstrap relation btw. the electron and phonons

$$H = -J \sum_{\langle \mathbf{n},\mathbf{m} \rangle} |\mathbf{n}\rangle\langle\mathbf{m}| + \sum_{\mathbf{n}} \left[\hbar\omega_0 b_{\mathbf{n}}^\dagger b_{\mathbf{n}} + \gamma |\mathbf{n}\rangle\langle\mathbf{n}| \left(b_{\mathbf{n}} + b_{\mathbf{n}}^\dagger \right) \right]$$

- a genuine many-body problem
  - 1 electron + multiphonon cloud moving with it
- perturbation expansion from two limiting cases
  - free-electron limit ($\gamma = 0$); atomic limit ($J = 0$)
- 2d materials, organic semiconductors, photosynthetic pigment–protein complexes
  - intermediate regime: $J \sim \hbar\omega_0 \sim \gamma \sim k_B T$

Polaron problem: An old yet not fully solved problem

- ground state and thermodynamics @ finite T: OK
- single-particle Green's function, spectral properties: ???
  - especially @ finite T
- many **approximate** techniques (numerically **cheap** & physically **transparent**)
  - canonical transformations (*à la* Lang–Firsov)
  - coherent-state expansion (*à la* Davydov)
  - cluster perturbation theory
  - momentum-average approximation
  - dynamical mean-field theory
- **numerically exact** techniques (computationally **intensive** & **less** physically **transparent**)
  - exact diagonalization (ED)
  - quantum Monte Carlo (QMC)
  - variational Hilbert space + finite-T Lanczos
  - finite-T density matrix renormalization group (DMRG)

Polaron problem: Numerically exact techniques

- the solution has converged wrt. the control parameter(s)
  - ED: number of excited phonons in each mode
  - QMC: Trotter discretization or diagram order or ...
- QMC is usually formulated in imaginary time
  - analytical continuation from the Matsubara axis
- finite-T Lanczos is formulated directly in real frequency
  - artificial broadening $\delta(\omega - \Omega_k) \to \frac{1}{\pi}\frac{\eta}{(\omega-\Omega_k)^2+\eta^2}$
    - J. Bonča *et al.*, Phys. Rev. B **100**, 094307 ('19).
- finite-T DMRG: real time, algorithmically complex
  - D. Jansen *et al.*, Phys. Rev. B **102**, 165155 ('20).
- a less complicated real-time method
  - chemical physics, open quantum systems: hierarchical equations of motion (HEOM)
    - review: Y. Tanimura, J. Chem. Phys. **153**, 020901 ('20).
  - system of interest: the electron; environment: phonons
  - trace out/integrate over phonons

Feynman–Vernon influence functional theory

- usually in real space, here in the momentum space
  - R. P. Feynman and F. L. Vernon, Ann. Phys. (NY) **24**, 118 ('63).

Greater Green's function and its influence phase

$$G^{>}(k,t) = -i\langle c_k(t)c_k^\dagger(0)\rangle = -ie^{-i\Omega_k t}\langle k|\rho_k^{(I,>)}(t)|\mathrm{vac}\rangle$$

$$\langle k|\rho_k^{(I,>)}(t)|\mathrm{vac}\rangle = \mathcal{U}^{(I,>)}(t)|k\rangle\langle\mathrm{vac}|$$

$$\mathcal{U}^{(I,>)}(t) = T\exp[-\Phi^{(I,>)}(t)]$$

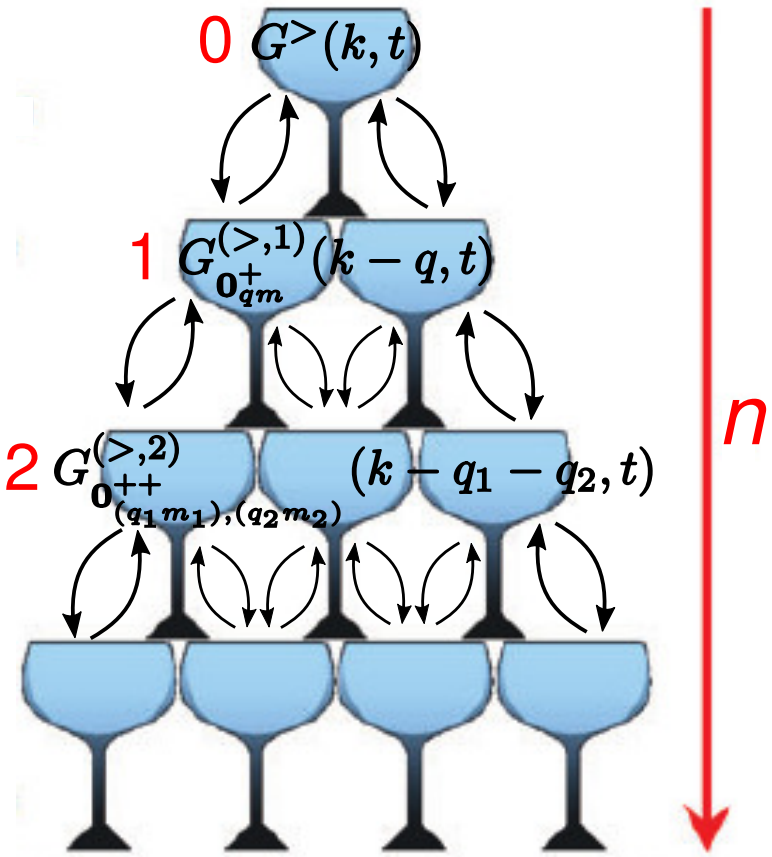
$$\Phi^{(I,>)}(t) = \omega_0^2\int_0^t ds_2\int_0^{s_2} ds_1 \sum_{qm} V_q^{(I)}(s_2)c_{qm}e^{-\mu_{qm}(s_2-s_1)}V_{-q}^{(I)}(s_1)$$

$$V_q = \sum_k |k+q\rangle\langle k|$$

$$m = 0,1;\ \mu_{qm} = (-1)^m i\omega_0;\ c_{qm} \propto \gamma^2\left(\frac{1+(-1)^m}{2} + N_{\mathrm{ph}}\right)$$

V. J. & N. V., Phys. Rev. B **105**, 054311 ('22).

Exact (Feynman–Vernon) → Numerically exact (HEOM)



HEOM for $N=1$ and $D=3$

V. J. & N. V., Phys. Rev. B **105**, 054311 ('22).

- $G^{(>,n)}_{\mathbf{n}}(k-k_{\mathbf{n}},t)$
  - $n \geq 0$: no. of scatterings
  - $\mathbf{n}$: all phonon wave vectors $\mathbf{q}$, emission ($m=0$), absorption ($m=1$)
  - $k_{\mathbf{n}}$: net momentum exchange after n scatterings
- hierarchical links $n \leftrightarrow n \pm 1$
- truncation at $n = D$
  - $n_{\mathrm{AGF}} = \binom{2N+D}{D}$
  - dangling bonds
- errors: finite N and D
- $A(k,\omega) = -\frac{1}{2\pi}\mathrm{Im}\, G^{>}(k,\omega)$
- 1d system

QMC method: consistency check on the imaginary axis

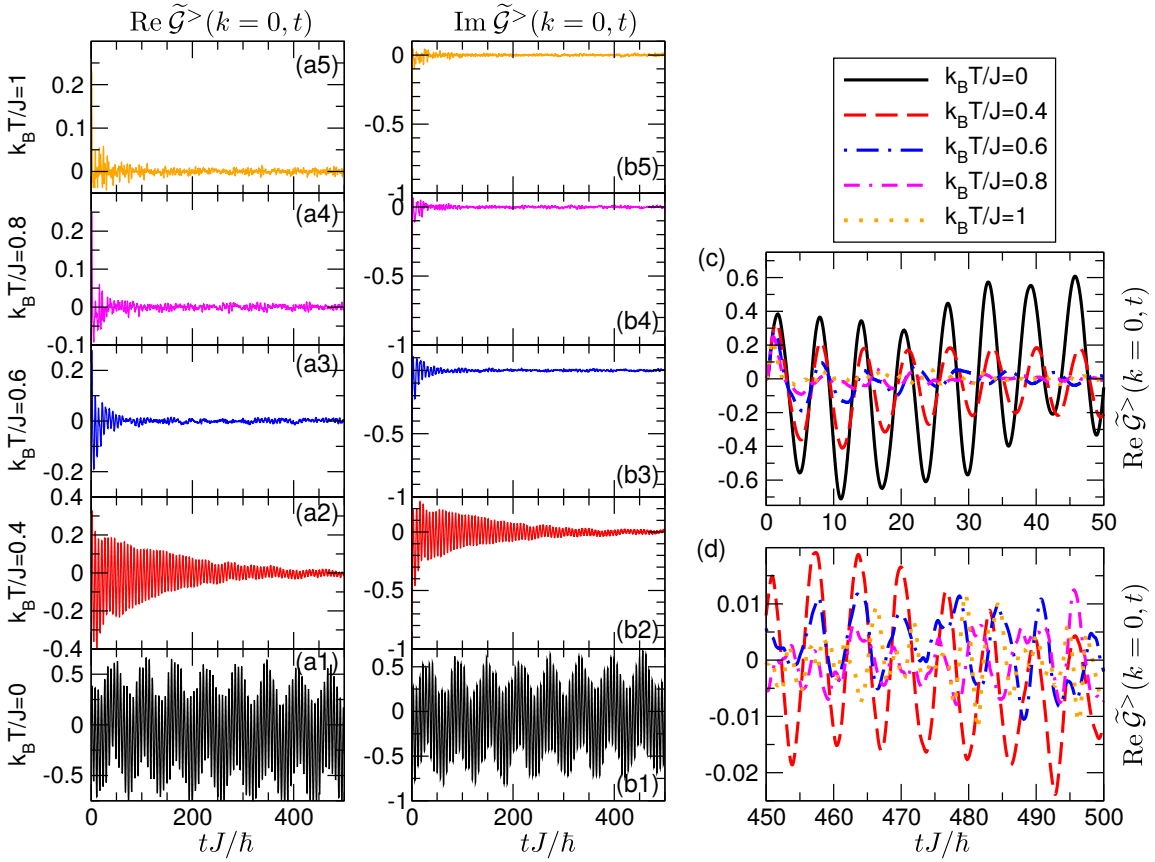
- $C(k,\tau) = \left\langle c_k(\tau) c_k^\dagger \right\rangle$, $0 \leq \tau \leq \beta\hbar$
  - path-integral representation
  - imaginary-time discretization (Suzuki–Trotter)
  - **phonons** are integrated out **analytically**
  - **direct sampling** of the **electronic** phase space, **no** need for **Metropolis** algorithm
- extension of early works dealing with thermodynamic quantities only
  - De Raedt and Lagendijk, Phys. Rev. Lett. **49**, 1522 ('82).
  - De Raedt and Lagendijk, Phys. Rev. B **27**, 6097 ('83).
- $C_{\mathrm{HEOM}}(k,\tau) = \int d\omega\, e^{-\omega\tau} A(k,\omega)$

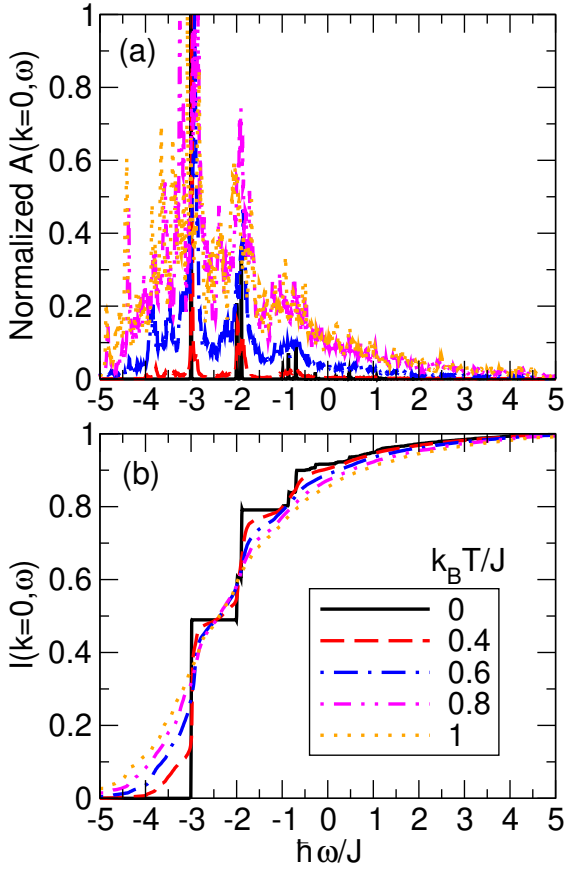
V. J. & N. V., Phys. Rev. B **105**, 054311 ('22).

Results (1): $\gamma/J = \sqrt{2}$, $\hbar\omega_0/J = 1$, $N = 8$, $D = 7$

crossover (large → small polaron) and extreme quantum regime

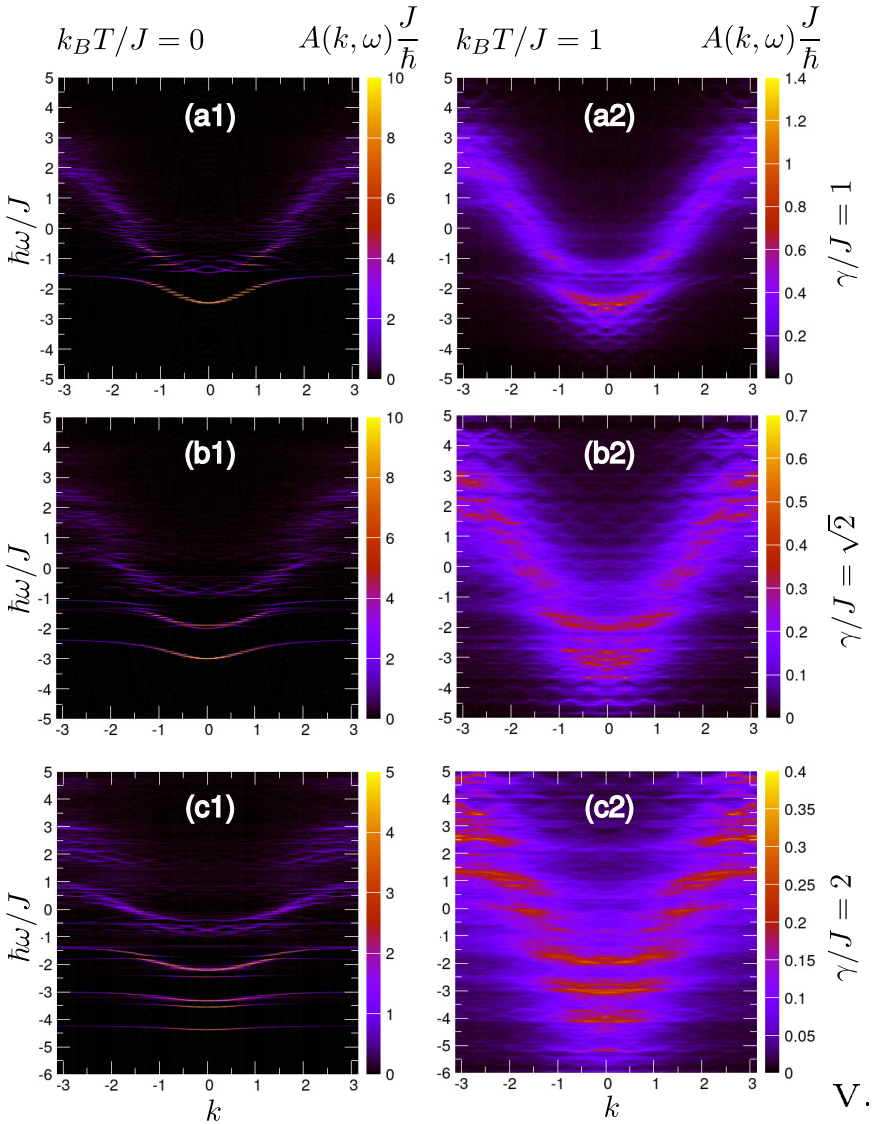
$$G^{>}(k,t) = e^{-i\Omega_k t} \times \widetilde{\mathcal{G}}^{>}(k,t) \qquad I(k,\omega) = \int_{-\infty}^{\omega} d\omega' A(k,\omega')$$



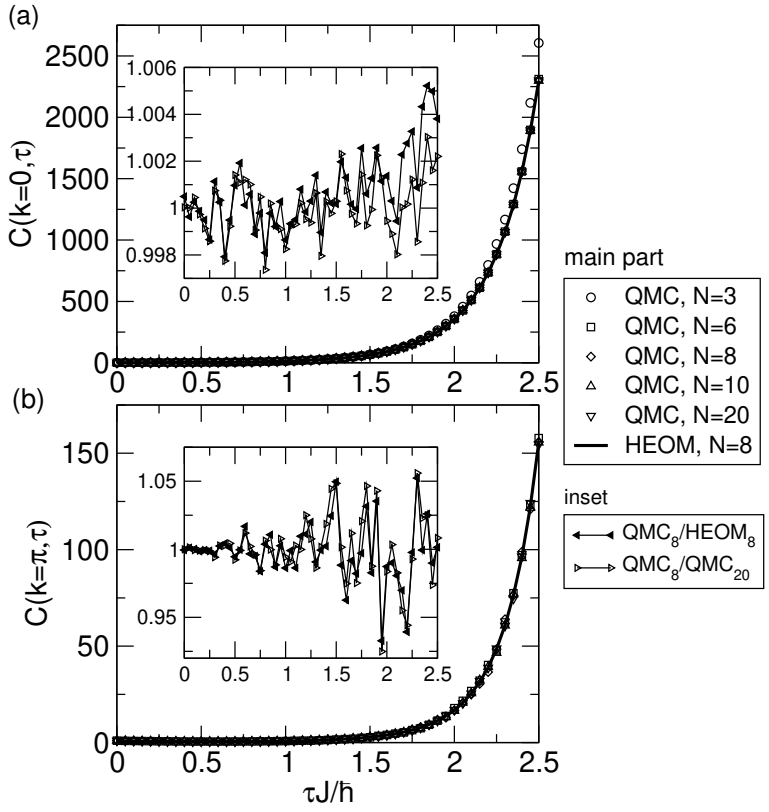


V. J. & N. V., Phys. Rev. B **105**, 054311 ('22).

Results (2): $\hbar\omega_0/J = 1$



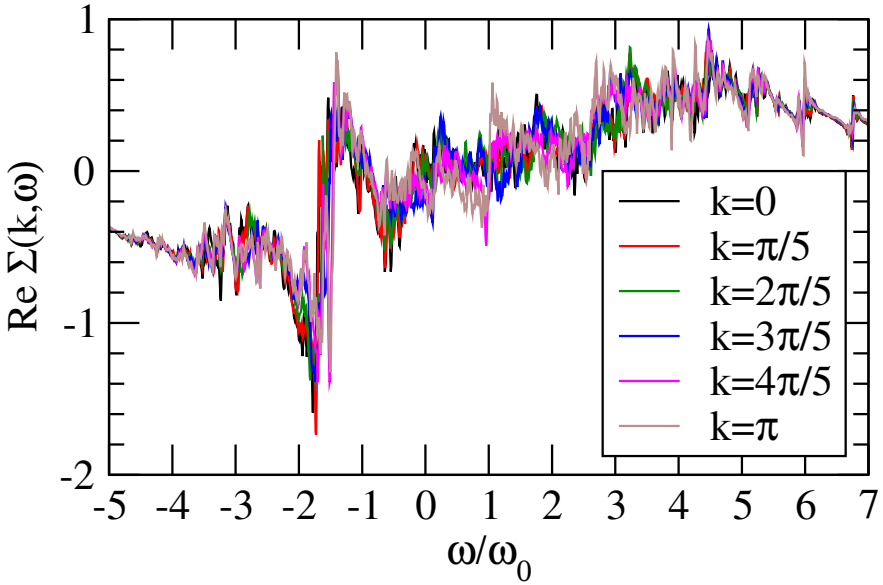
$\gamma/J = \sqrt{2}$, $k_BT/J = 0.4$

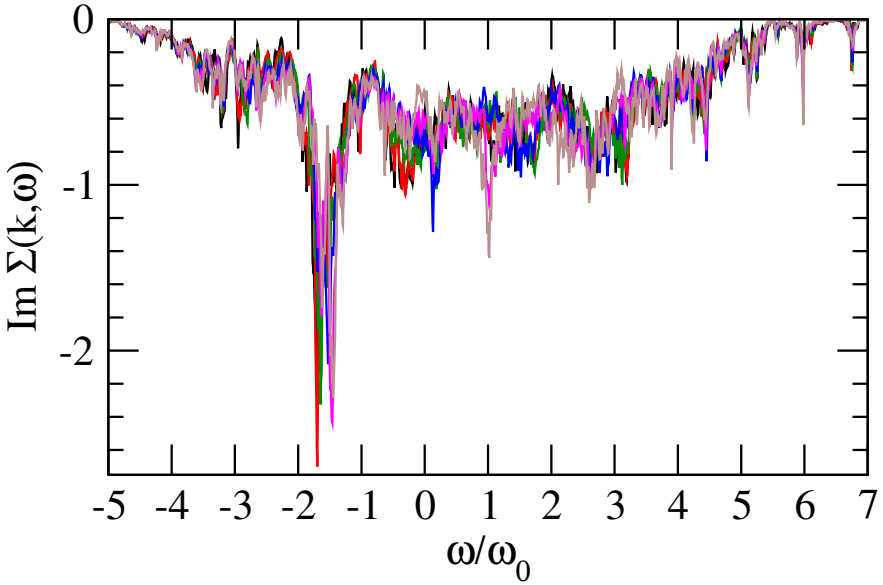


V. J. & N. V., Phys. Rev. B **105**, 054311 ('22).

Self energy $\Sigma(k,\omega)$ — weak nonlocality of correlations

- $\gamma/J = \hbar\omega_0/J = 1$, $k_B T/J = 0.7$; $N = 10$, $D = 6$





- $\Sigma(k,\omega)$ quite weakly depends on k
- correlations are predominantly local
- what about DMFT? → **the next talk by P. Mitrić**

Epilogue

- HEOM is a promising approach to study finite-T dynamical properties of the Holstein model
- advantage: real-time formulation
- disadvantage: number of auxiliaries grows combinatorially with N and D
  - a new numerical implementation is required for higher-dimensional models
- all details in V. J. & N. V., Phys. Rev. B **105**, 054311 ('22).
- in 1d Holstein model, correlations are predominantly local
  - DMFT as a numerically cheap alternative? **next talk**