Photoemission spectra and optical response of many-polaron systems

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Motivation

- Optical measurements have proven the importance of electron-phonon (EP) coupling and even polaron effects in important classes of materials, including quasi-1d MX chains, quasi-2d cuprate superconductors, and 3d colossal magnetoresistance manganites.
- In all these systems, a noticeable density of (polaronic) charge carriers is observed, which puts the applicability of single-polaron theories into question, particularly in the often realized case of intermediate EP couplings and phonon frequencies.
- Questions: What happens to polarons if their density is large enough so that individual quasiparticles (QP) would overlap? Is there a density-driven crossover from (large) polarons to weakly dressed electrons?
- Problem: Even for the most simplified models no reliable analytical results exist for finite carrier densities in the intermediate EP coupling regime!

Model

1D spinless fermion Holstein model:

$$H = -t\sum_{\langle i,j\rangle} c_i^\dagger c_j - g\,\omega_0 \sum_i (b_i^\dagger + b_i) n_i + \omega_0 \sum_i b_i^\dagger b_i$$

Parameter ratios:

$$g^2 = \varepsilon_p/\omega_0 \quad ; \quad \lambda = \varepsilon_p/2t$$
$$\alpha = \omega_0/t \quad ; \quad n = N_e/N$$

Known results:

- For a single particle a transition from a large polaron to a small polaron takes place with increasing EP coupling strength (at least in 1D).
- In the intermediate coupling regime $\lambda \simeq 1$, $g^2 \simeq 1$, the size of the polaron is strongly dependent on the phonon frequency:

  $\alpha \ll 1$: rather extended distortion

  $\alpha \gg 1$: localised distortion.
- Focusing on the intermediate coupling adiabatic regime we expect strong density-effects due to a possible overlap of phonon clouds (of course, in the $U \to \infty$ limit, bipolaron formation is suppressed)!

Methods

Quantum Monte Carlo (QMC) [1,2]

- Grand-canonical approach, free of autocorrelations.
- Finite-temperature Trotter discretization ($\Delta\tau = 0.1$).
- Lang-Firsov transformed model $\rightsquigarrow$ moderate sign problem [which is most pronounced for intermediate λ and small α at low temperatures T ($\beta = 1/k_{\rm B}T$)].
- Maximum entropy method to get dynamic quantities.

Exact diagonalization (ED) [3]

- Lanczos technique $H^D \to T^L$ (Krylov subspaces): Fast convergence for extremal eigenvalues E_0, $|\psi_0\rangle$ ($D \lesssim 10^{11}$, $L = 100 \rightsquigarrow \Delta E_0 \lesssim 10^{-9}$)!
- Phonons? $D = \infty$! $\rightsquigarrow$ Controlled Hilbert space truncation combined with a density-matrix based phonon basis optimisation procedure.

Dynamical properties at T=0?

$$A^{\mathcal{O}}(\omega) = -\frac{1}{\pi}\lim_{\eta\to 0}\left\langle \psi_0 \left| \mathcal{O}^\dagger \frac{1}{\omega - H + E_0 + {\rm i}\eta}\mathcal{O}\right| \psi_0\right\rangle = \sum_n |\langle \psi_n|\mathcal{O}|\psi_0\rangle|^2 \delta[\omega - (E_n - E_0)]$$

$\hookrightarrow$ complete spectrum necessary!?

Kernel polynomial method (KPM) [4]

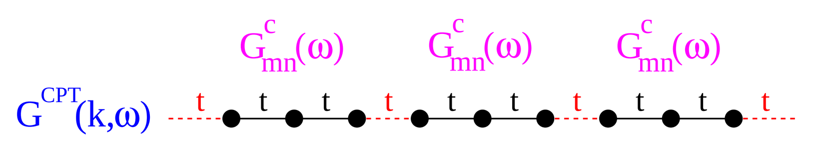
- Expansion of $\delta[\ldots]$ using Chebyshev polynomials
$$A^{\mathcal{O}}(x) = \frac{1}{\pi\sqrt{1-x^2}}\left(\mu_0^{\mathcal{O}} + 2\sum_{m=1}^{\infty}\mu_m^{\mathcal{O}} T_m(x)\right)$$
- Determination of moments
$$\mu_m^{\mathcal{O}} = \int_{-1}^{1} dx\, T_m(x) A^{\mathcal{O}}(x) = \langle \psi_0|\mathcal{O}^\dagger T_m(X)\mathcal{O}|\psi_0\rangle$$
by iterative MVM ($X = (H-b)/a \to E_n \in [-1,1]$)
- Problem: $M < \infty$! $\rightsquigarrow$ Gibbs oscillations $\rightsquigarrow$ truncation errors! Solution: Damping factors (e.g., Jackson or Lorentz kernels)!
- (FFT) Reconstruction of $A^{\mathcal{O}}(x)$ from the M calculated moments via linear approximation (KPM) or nonlinear optimisation procedure (MEM).

Advantages of KPM:

- High-resolution applications – polarons!
- Uniform reconstruction of spectra – gap features)!
- CPU-time ($\propto MD$); trace – average over random $|r\rangle$.

Cluster Perturbation Theory (CPT) [5]

Green function $G(k,\omega)$ on an infinite lattice ($N_c = \infty$)?



- We have: Green function $G^c_{mn}(\omega)$ on finite cluster(s) of N_c sites (OBC) !
- 1st order perturbation in $V = \sum \left[-\frac{t}{\ }-\right]$

$$G^{(1)}_{ij}(\omega) = G^c_{ij}(\omega) + \sum_{rs} G^c_{ir}(\omega) V_{rs} G^{(1)}_{sj}(\omega)$$

$$G^{(1)}_{mn}(K,\omega) = \left(\frac{G^c(\omega)}{1 - V(K)G^c(\omega)}\right)_{mn}, \quad (K = N_c k)$$

- Fourier transform:

$$G^{\rm CPT}(k,\omega) = \frac{1}{N_c}\sum_{m,n=1}^{N_c} G^{(1)}_{mn}(K,\omega)e^{-{\rm i}k(m-n)}$$

Observables

- Single-particle spectral function:

$$A^{\pm}_k(\omega) = \sum_m |\langle m^{(N_{el}\pm 1)}|c^{\pm}_k|0^{(N_{el})}\rangle|^2 \times \delta[\omega \mp (E^{(N_{el}\pm 1)}_m - E^{(N_{el})}_0)]$$

with $c^+_k = c^\dagger_k$, $c^-_k = c_k$.

$A^-_k(\omega)$ [$A^+_k(\omega)$] is related to the [inverse (I)] photo-emission (PE) of an electron.

Note that $A_k(\omega-\mu) = -\frac{1}{\pi}{\rm Im}G(k,\omega-\mu)$, where the imaginary-time Green function $G(k,\tau)$ can be directly measured by QMC.

- Partial densities of states (DOS):

$$\rho^{\pm}(\omega) = \sum_k A^{\pm}_k(\omega)\,.$$

- Optical conductivity, ${\rm Re}\,\sigma(\omega) = D\delta(\omega) + \sigma^{\rm reg}(\omega)$, the regular part of which is given by

$$\sigma^{\rm reg}(\omega) = \frac{\pi}{N}\sum_{n>0}\frac{|\langle n|\hat{\jmath}|0\rangle|^2}{E_0 - E_n}\delta(\omega - (E_0 - E_n))\,,$$

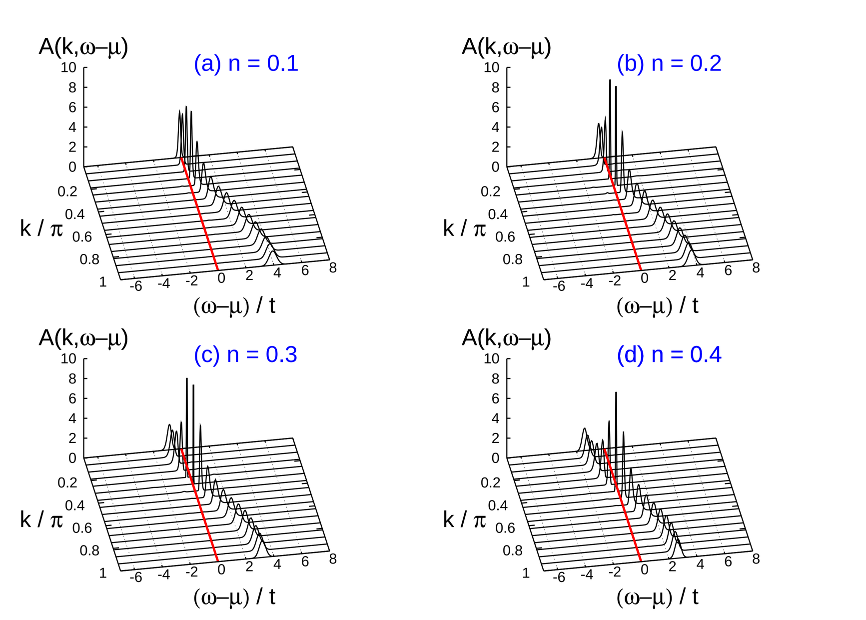
where $\hat{\jmath} = {\rm i}et\sum_i (c^\dagger_i c_{i+1} - c^\dagger_{i+1}c_i)$ (current operator).

Numerical results

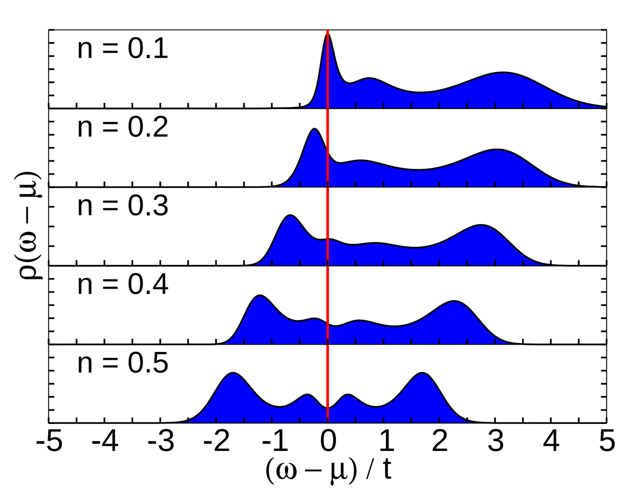
Weak coupling

$\lambda = 0.1$, $\alpha = 0.4$ – QMC results for $N = 32$, $\beta t = 8$

single-particle spectral function (QMC)



density of states



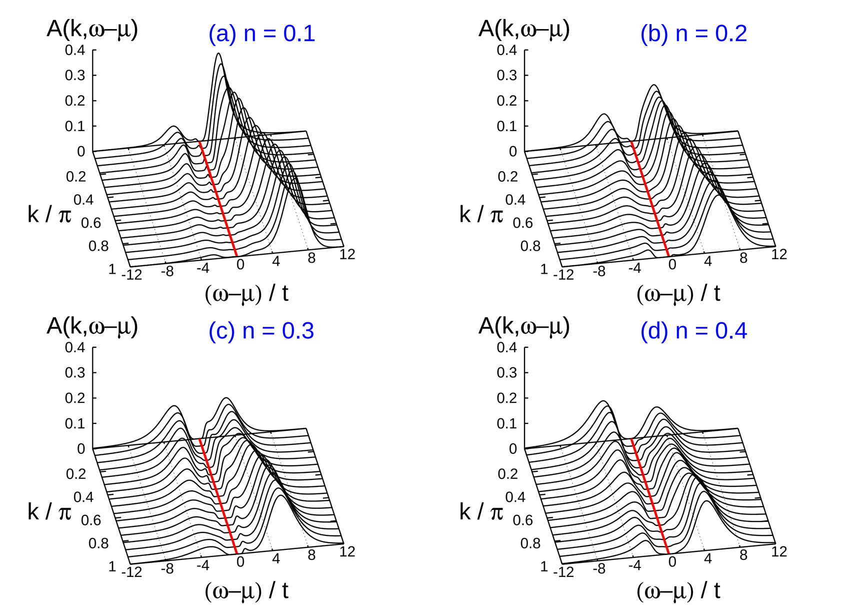
- pronounced QP peak $\forall n$
- gap feature develops for $n \to 0.5 \Leftrightarrow$ CDW

$\hookrightarrow$ dressed "electronic" QPs!

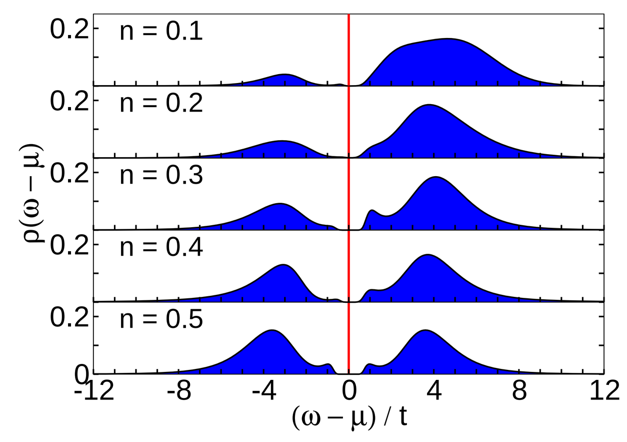
Strong coupling

$\lambda = 2.0$, $\alpha = 0.4$ – QMC results for $N = 32$, $\beta t = 8$

single-particle spectral function (QMC)



density of states



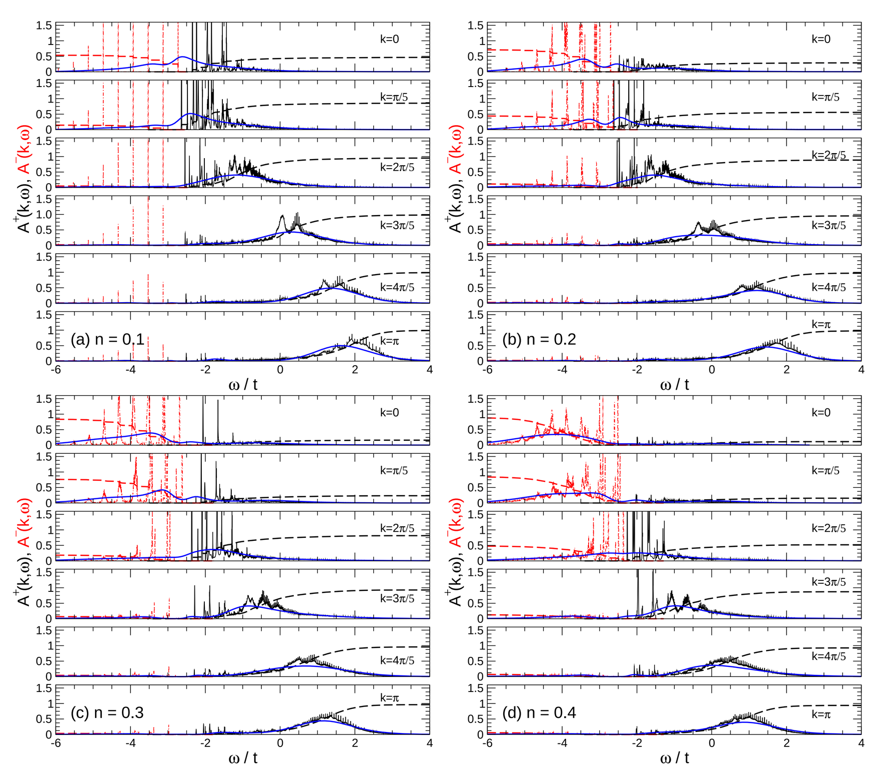
- exponentially small spectral weight at μ $\forall n$
- QP band - "gap" - broad incoherent feature

$\hookrightarrow$ small "polaronic" QPs!

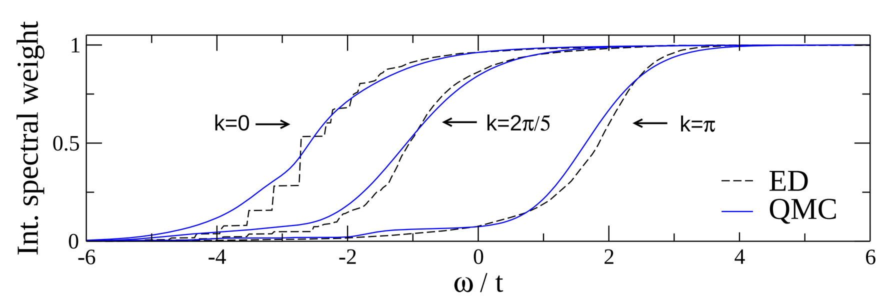
Intermediate coupling

$\lambda = 1.0$, $\alpha = 0.4$, $N = 10$ (ED) and $N = 10$, $\beta t = 8$ (QMC)

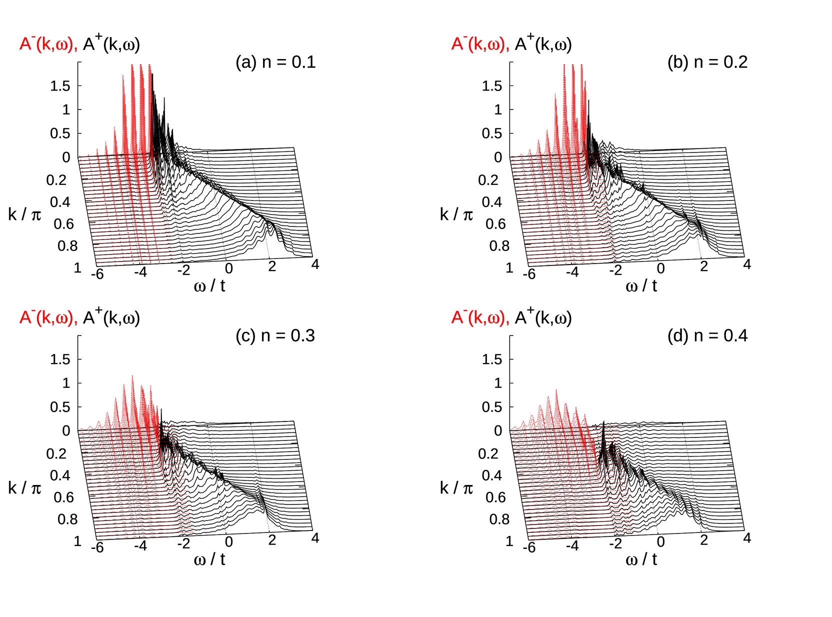
single-particle spectral function (ED)



integrated spectral weight (QMC vs. ED)



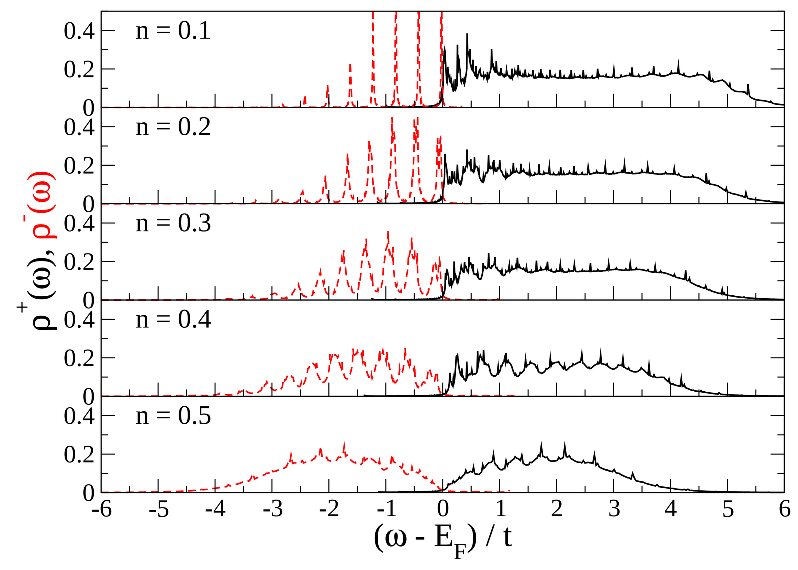
spectral functions (CPT)



- $n = 0.1$: Fermi level lies within the polaron band; band flattening at large wave-vectors k.
- Incoherent part of the spectrum closely follows the free electron (cosine) dispersion.
- Polaron band merges with incoherent excitations at about $n = 0.3 \rightsquigarrow$ no clear separation of the QP peak!

$\hookrightarrow$ density-driven change of the "character" of the charge carriers!

density of states (CPT)

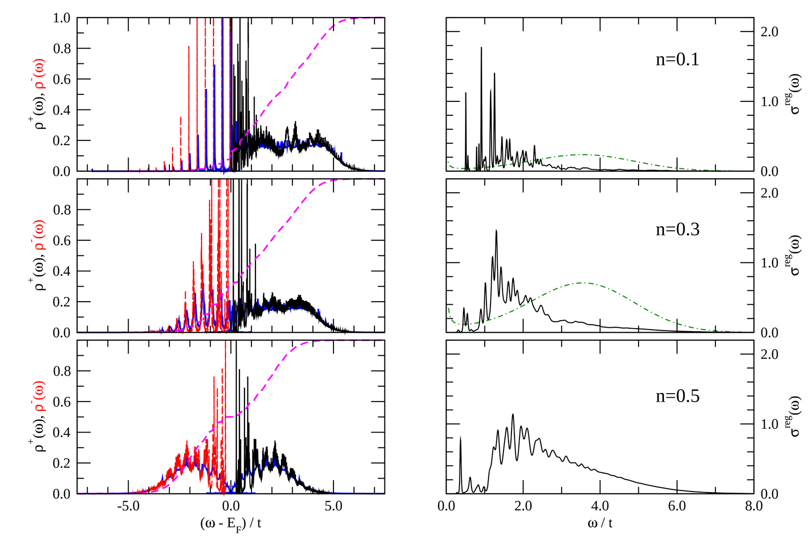


- $n \nearrow$: small (polaronic) DOS at $E_{\rm F}$ ($\propto {\rm e}^{-g^2}$) $\to$ "metallic" DOS at $E_{\rm F}$ ("polaron dissociation") $\to$ pseudo-gap - precursor of CDW ($\exists$ for $\lambda > \lambda_c(\omega_0)$)

$\hookrightarrow$ crossover from polaronic to metallic behaviour!

partial DOS & optical response (ED)

$\lambda = 1.0$, $\alpha = 0.4$, i.e., $g^2 = 5$, $N = 10$



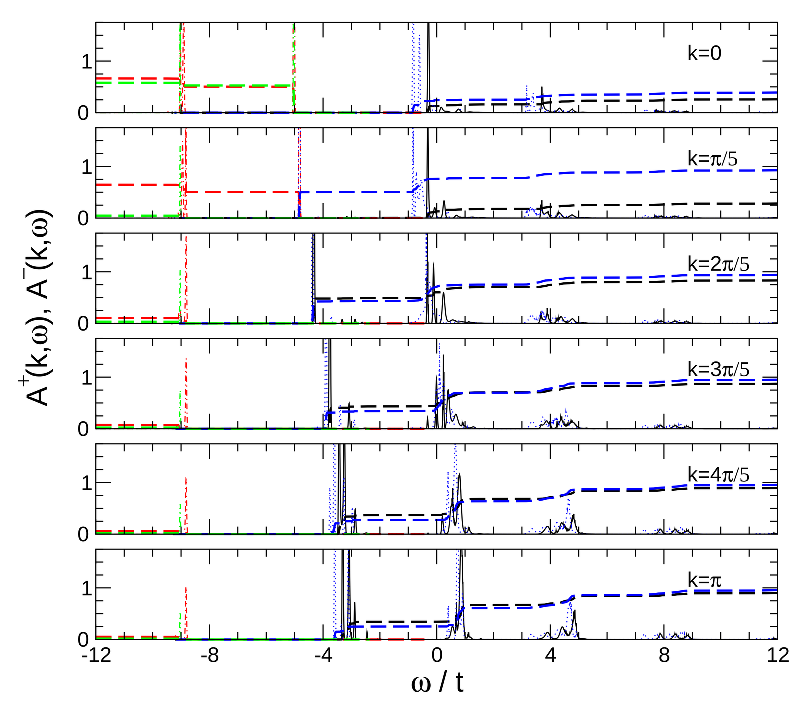
For comparison, the analytical strong coupling result [6]

$$\sigma^{\rm reg}(\omega) = \frac{\sigma_0 n}{\omega_0 g\omega}\exp\left[\frac{(\omega - 2g^2\omega_0)^2}{4g^2\omega_0^2}\right],$$

is included ($\sigma_0 = 8$) – noticable deviations!.

Anti-adiabatic (strong-coupling) regime

$\lambda = 2.0$, $\alpha = 4.0$, $N = 10$ (ED)



- Phonon absorption bands with electronic satellites.
- Spectra are mainly unchanged going from $n = 0.1$ [green/blue] to $n = 0.3$ [red/black] (i.e., ther are almost no finite-density effects).

$\hookrightarrow$ Carriers are small polarons also at large fillings!

Conclusions

In the physically most important adiabatic intermediate electron-phonon coupling regime, for which no analytical results are available, we observe a dissociation of polarons with increasing band filling, leading to normal metallic behaviour, while for parameters favouring small polarons, no such density-driven changes occur. The present work points towards the inadequacy of single-polaron theories for a number of polaronic materials such as the manganites.

[1] Hohenadler, Evertz, von der Linden, Phys. Rev. B **69**, 024301 (2004).

[2] Hohenadler et al., Phys. Rev. B **71**, 245111 (2005).

[3] Bäuml, Wellein, Fehske, Phys. Rev. B **58**, 3663 (1998);
Weiße et al., Phys. Rev. B **62**, R747 (2000).

[4] Weiße et al., cond-mat/0504627 (review).

[5] Sénéchal, Perez, Plouffe, Phys. Rev. B **66**, 075129 (2002);
Hohenadler, Aichhorn, von der Linden, Phys. Rev. B **68**, 184304 (2003).

[6] D. Emin, Phys. Rev. B **48**, 13691 (1993).