Spectral signatures of the Luttinger liquid to charge-density-wave transition

M. Hohenadler, G. Wellein, A. R. Bishop, A. Alvermann, and H. Fehske

1Institute for Theoretical and Computational Physics, TU Graz, 8010 Graz, Austria
2Computing Center, University Erlangen, 91058 Erlangen, Germany
3Theoretical Division and Center for Nonlinear Studies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
4Institute for Physics, Ernst-Moritz-Arndt University Greifswald, 17487 Greifswald, Germany

Electron–phonon spectral functions of the one-dimensional, spinless-fermion Holstein model at half filling are calculated in the four distinct regimes of the phase diagram, corresponding to an attractive or repulsive Luttinger liquid at weak electron-phonon coupling, and a band–or polaronic insulator at strong coupling. The results obtained by means of kernel polynomial and systematic cluster approaches reveal substantially different electronic correlations, has renewed the interest in models of the strong-EP coupling regime and in conjunction with strong metallic complexes, ferroelectric perovskites, conjugated polymers, or organic charge-transfer salts are very susceptible to structural distortions driven by electron-phonon (EP) interaction. At commensurate band fillings these systems might undergo a Peierls or charge-density-wave (CDW) instability accompanied by a dimerization of the lattice but, unlike in conventional metals, both quantum lattice fluctuations and strong electronic correlations are important. The challenge of understanding the related metal-insulator transition, especially in the strong-EP coupling regime and in conjunction with strong electronic correlations, has renewed the interest in models of interacting electrons and phonons.

In this work, we compare in detail the spectral signatures of four physically distinct regimes in the phase diagram of the one-dimensional (1D) Holstein model of spinless fermions (HMSF) (Fig. 1). To this end, we calculate the single-particle spectral functions of electrons and phonons, using exact diagonalization (ED) in combination with cluster perturbation theory (CPT) and the kernel polynomial method (KPM). Besides the influence of the degree of phonon retardation on the nature of the Peierls transition, the wave-number dependence of the spectral functions missed in previous dynamical mean-field theory studies turns out to be crucial for the 1D case considered here.

The paper is organized as follows. The model is introduced and its phase diagram reviewed in Sec. II. In Sec. III we present the methods used, and results are discussed in Sec. IV. Finally, we summarize in Sec. V.

II. MODEL

The Hamiltonian

\[ H = -t \sum_{\langle i,j \rangle} c_i^\dagger c_j + \omega_0 \sum_i b_i^\dagger b_i - g \omega_0 \sum_i \hat{n}_i (b_i^\dagger + b_i), \] (1)

Despite its simplicity, the HMSF is not exactly solvable and a wide range of analytical and numerical methods have been applied to map out the ground-state phase diagram in the \( g, \omega_0 \) plane (see references in Ref. [8]). At present the probably most precise phase boundary is obtained by exact diagonalization and density matrix renormalization group (DMRG) techniques. More recent large-scale DMRG calculations supplemented by a finite-size analysis have proved that at low (high) phonon frequencies the metallic LL phase is characterised by an attractive (repulsive) interaction. But also above \( g_c(\omega_0) \), where long-range CDW order sets in, exist two physically distinct regimes, which can be classified, e.g., by their different optical response, either as a band insulator in the adiabatic regime \( \omega_0/t \ll 1 \) or as a polaronic superlattice in the limit of large phonon frequencies \( \omega_0/t \gg 1 \) (see Fig 1). Including the spin degrees of freedom and a finite Hubbard interaction allows for additional quantum phase transitions between Peierls and Mott insulating phases. The half-filled HMSF captures the relevant physics of the more general quarter-filled Hubbard-Holstein model in the regime of large Hubbard repulsion \( U \gg t \), often realized in experiment, where on-site bipolaron formation is suppressed.
FIG. 1: DMRG ground-state phase diagram of the 1D half-filled HMSF (cf. Refs. 2,9). In the adiabatic limit $\omega_0 \to 0$ the critical dimensionless coupling constant $\lambda_c$ converges to zero. For $\omega_0 > 0$, due to quantum phonon fluctuations, there exists a finite critical coupling $g_c(\omega_0)$. For $2\omega_0 \to \infty$, the model exhibits a Kosterlitz-Thouless phase transition near $g_c^* = g_c(\omega_0 \to \infty)$. Crosses indicate parameter sets used below.

III. METHODS

The $T = 0$ electron spectral function is related to the one-electron Green function via

$$A(k, \omega) = -\frac{1}{\pi} \text{Im} \ G(k, \omega) = A^+(k, \omega) + A^-(k, \omega), \quad (2)$$

where

$$A^\pm(k, \omega) = -\frac{1}{\pi} \lim_{q \to 0^+} \langle \psi_0 | \hat{c}_k^\pm - \frac{1}{\omega + i\eta + \mathbf{H}} \hat{c}_k^\mp | \psi_0 \rangle, \quad (3)$$

$\hat{c}_k^\pm = c_k + c_k^\dagger$ and $| \psi_0 \rangle$ is the ground state of the HMSF with $N_c = N/2$ electrons. $A^-(k, \omega)$ [$A^+(k, \omega)$] describes (inverse) photoemission of an (injected) electron with momentum $k$ and energy $\omega$.

Relying on approximation-free numerical approaches (here ED) we can calculate exact Green functions for finite systems only. To obtain an approximation of $G(k, \omega)$ for the infinite lattice, we can exploit CPT. To this end, we divide the infinite system into identical clusters of $N_c$ sites each, and determine the electron cluster Green function $G_{ij}^{(c)}(\omega)$ for all non-equivalent pairs of sites $i, j = 1, \ldots, N_c$ by the KPM (for details see Ref. 7). The phonon Hilbert space is truncated such that the resulting error of the spectra is negligible ($< 10^{-4}$), and we have used 1024 Chebyshev moments. In a second step, the Green function $G(k, \omega)$ for the infinite lattice is obtained from the first-order result

$$\tilde{G}_{ij}(k, \omega) = \left[ \frac{G^{(c)}(\omega)}{1 - t(k) G^{(c)}(\omega)} \right]_{ij}$$

of a strong-coupling expansion in the inter-cluster hopping operator $\tilde{t}(k)$ as

$$G(k, \omega) = \frac{1}{N_c} \sum_{i,j=1}^{N_c} \tilde{G}_{ij}(k, \omega) e^{-ik\cdot(r_i-r_j)}. \quad (5)$$

The $T = 0$ phonon spectral function is defined as

$$B(q, \omega) = -\frac{1}{\pi} \text{Im} \ D(q, \omega) \quad (6)$$

with

$$D(q, \omega) = \lim_{\eta \to 0^+} \langle \psi_0 | \hat{\pi}_q \frac{1}{\omega + i\eta + \mathbf{H}} \hat{\pi}_{-q} | \psi_0 \rangle \quad (7)$$

for $\omega \geq 0$ and $\hat{\pi}_q = N^{-1/2} \sum_j \hat{x}_j e^{-iq \cdot r_j}$. For the HMSF $\tilde{1}$, $B(q, \omega)$ is symmetric in $q$, and we have a dispersionless bare propagator $D_0(q, \omega) = 2\omega_0/(\omega^2 - \omega_0^2)$. EP interaction will renormalize the phonon frequency, whereby $D(q, \omega)$ attains a $q$-dependence. Concerning notation, we shall reserve the capital $Q$ for allowed wavenumbers of finite clusters.

IV. RESULTS

The crosses in the phase diagram (Fig. 1) mark the four parameter sets to be used in the sequel.

Figures 2(a) and (c) show results for the CPT electron spectral functions $A^\pm(k, \omega)$ in the metallic region. In the adiabatic (attractive LL) regime [Fig. 2(a)], we find a rather pronounced peak at the Fermi level $E_F$ but nevertheless the system is not a Fermi liquid. The non-universal LL parameters (charge velocity and interaction coefficient) can be determined from a DMRG finite-size scaling. At $k = k_F$, very little spectral weight is contained in the incoherent part of the spectrum. By contrast, away from the Fermi momentum, almost all of the spectral weight resides in the incoherent part, whose maximum follows quite closely the free dispersion $-2t \cos k$ [notice the symmetry of $A^- (k < k_F)$ and $A^+ (k > k_F)$]. As these states are accessible only via (multi–) phonon excitations, the width of the incoherent band is proportional to $g^2$.

The spectrum in the anti-adiabatic (repulsive LL) regime, Fig. 2(c), looks significantly different. Due to the larger coupling $\lambda = \epsilon_p/2t = 1$ (with respect to the bare electronic bandwidth), as compared to $\lambda = 0.5$ in Fig. 2(a), the carriers are more strongly renormalized. This is reflected in the reduced bandwidth $W \approx 3t$, with the spectral weight being more evenly distributed than in the adiabatic case. The fact that $\omega_0 > W$ gives rise to multiple, non-dispersive side-bands at energies $E_F \pm n\omega_0$.

In Figs. 2(b) and (d) we present the corresponding phonon spectral functions, as obtained from ED. Again we find pronounced differences between the adiabatic [Fig. 2(b)] and the anti-adiabatic [Fig. 2(d)] cases, revealing the distinct nature of the Peierls instability—driven by $\lambda$ or $g$—for small and large $\omega_0/t$.

For $\omega_0/t = 0.4$ [Fig. 2(c)], we observe a peak at $\omega = 0$ originating from the homogeneous ($Q = 0$) shift of the electronic level for $\lambda > 0$, as well as a signal located at the bare
FIG. 2: (Color online) Left panel [(a),(c)]: Spectral density for photoemission \([A^{-}(k, \omega); \text{red dashed lines}]\) and inverse photoemission \([A^{+}(k, \omega); \text{black solid lines}]\) from CPT in the metallic LL phase. Energies are measured relative to the Fermi level \(E_{F}\). Right panel [(b),(d)]: Corresponding exact phonon spectral function \(B(Q, \omega)\) for the allowed wave-numbers \(Q\) of \(N\)-site clusters. Electron and phonon spectra have been calculated in the adiabatic [attractive LL; (a),(b)] and anti-adiabatic [repulsive LL; (c),(d)] weak-to-intermediate EP coupling regimes. Dashed vertical lines in (b), (d) indicate the bare phonon frequency.

phonon energy \(\omega_0\). More importantly, the phonon excitations gradually soften near the zone boundary \((Q = \pi)\) already in the LL phase, a behavior characteristic of the Peierls transition traditionally regarded as a displacive phase transition.

For large \(\omega_0/t\) [Fig. 2(d)], we observe two main absorption features. The first excitation near \(\omega_0\) becomes even harder with increasing wave number, in contrast to the phonon softening in the adiabatic regime. The second branch emerging from \(\omega = 0\) \((Q = 0)\) resembles the two-spinon continuum \(\mathbb{K}\) of the XXZ model (onto which the spinless Holstein model may be mapped in the anti-adiabatic strong-coupling limit \(\mathbb{K}\)), i.e., it can be traced back to phonon-signatures of the corresponding electronic excitations.

We now turn to the insulating state of the HMSF (Fig. 3). Due to the Peierls distortion, and the formation of long-range CDW order for \(N \rightarrow \infty\), a gap opens at \(E_{F}\). In the adiabatic regime, the transition is from a LL to a traditional Peierls band insulator, whereas in the anti-adiabatic regime, the charge carriers first undergo a cross-over to small-polarons (polaronic metal), which then (upon increasing the coupling further) order to form a polaronic superlattice.

From Fig. 3(a) we see that the gap in the single-particle spectrum is still quite small. This is partly due to the fact that CPT in the form used here does not fully take into account long-range order. At \(k_{F}\), the strongest signatures lie just above and below \(E_{F}\), with the incoherent part again being very small. Away from \(k_{F}\), the incoherent band is significantly broadened (again almost \(\propto g^2\) at \(k = \pi\)), and reflects the Poisson distribution of the phonons in the ground state. Nevertheless, we can still detect the dispersion of the split electronic band.

In contrast to the adiabatic case of Fig. 3(a), Fig. 3(c) shows a clear gap for all \(k\). The polaronic charge carriers—in addi-
The phonon spectra for the insulating phase are shown in Figs. 3(b),(d). In the adiabatic case of Fig. 3(b), the zone boundary phonon has almost completely softened. In the limit $N \to \infty$, we expect a perfect degeneracy of $Q = 0, \pi$ for $g > g_c$, and a macroscopic population of the soft phonon mode. For $\omega_0/t = 4$ [Fig. 3(d)], there exist two completely flat signatures at $\omega = 0$ and $\omega = \omega_0$ [cf. Fig. 4(d)], the former having very large spectral weight due to the large “phonon content” of the polaron band.

Similar to the results for $A(k, \omega)$, it is desirable to obtain results for the phonon spectrum of the infinite system. However, in deriving a CPT-like approach to the phonon Green function $D(q, \omega)$, it turns out that the first-order correction in $\tilde{t}$ vanishes identically, since the electron number per cluster is a conserved quantity. Therefore, CPT results in a simple Fourier transformation of the cluster Green function (again obtained by the KPM),

$$D(q, \omega) = \frac{1}{N_c} \sum_{i,j=1}^{N_c} D_{ij}^{(c)}(\omega) e^{-i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}. \quad (8)$$

Notice that this result still becomes exact for the free Green function $D_0(q, \omega)$, and in the limit of strong coupling and large phonon frequency, when $t$ is a small parameter. As the calculation of the second-order contribution is extremely demanding we have restricted ourselves to this expansion to illustrate the $q$-dependence of $B(q, \omega)$ in Fig. 4.

The density plots in Fig. 4 clearly summarize the differences between the adiabatic and anti-adiabatic regimes, and between the LL and CDW phases of the HMSF. In Fig. 4(a) we see the renormalized phonon dispersion $\tilde{\omega}(q)$, which softens with increasing EP coupling, leading to a degeneracy of excitations at $Q = 0, \pi$ at $g_c$ [see panel (b)]. The strong zero-energy absorption feature at $\pi/2$ is an artifact of the small cluster size and the open boundary conditions used in the CPT.
the attractive LL (\(\omega_0/t = 0.4, g^2 = 2.5\)), band insulator (\(\omega_0/t = 0.4, g^2 = 5\)), repulsive LL (\(\omega_0/t = 4, g^2 = 0.5\)), and polaron superlattice regimes (\(\omega_0/t = 4, g^2 = 5\)), respectively.

Thus, with increasing phonon frequency, we find a cross-over from a soft-mode (displacive) to a central-peak-like (order-disorder-type) phase transition, similar to the analysis of the spin-Peierls transition motivated by \(\text{CuGeO}_3\).

V. SUMMARY

By presenting highly-reliable numerical results for the electron- and phonon spectral function, we have identified four physically distinct regions in the phase diagram of the one-dimensional spinless Holstein model, characterized by important generic features such as attractive or repulsive Luttinger liquid behavior and phonon softening, which are highly relevant for low-dimensional condensed matter systems.

Acknowledgments

This work was supported by the FWF project No. P15834, the DFG through SPP1073, KONWIHR and HPC Europa. Work at Los Alamos is performed under the auspices of US-DOE. We would like to thank M. Aichhorn, E. Jeckelmann, J. Loos, and A. Weiße for useful discussion, and acknowledge generous computer time granted by the HLRN Berlin, LRZ Munich and HLR Stuttgart.

\* Electronic address: hohenadler@itp.tu-graz.ac.at

1 N. Tsuda, K. Nasu, A. Yanese, and K. Siratori, Electronic Conduction in Oxides (Springer-Verlag, Berlin, 1990); A. R. Bishop and B. I. Swanson, Los Alamos Sciences 21, 133 (1993).

2 R. J. Bursill, R. H. McKenzie, and C. J. Hamer, Phys. Rev. Lett. 80, 5607 (1998); A. Weiße and H. Fehske, Phys. Rev. B 58, 13526 (1998); E. Jeckelmann, C. Zhang, and S. R. White, Phys. Rev. B 60, 7950 (1999).

3 D. Meyer, A. C. Hewson, and R. Bulla, Phys. Rev. Lett. 89, 196401 (2002); M. Capone and S. Ciuchi, Phys. Rev. Lett. 91, 186405 (2003); P. Paci, M. Capone, E. Cappelluti, S. Ciuchi, C. Grimaldi, and L. Pietronero, Phys. Rev. Lett. 94, 036406 (2005); G. Sangiovanni, M. Capone, C. Castellani, and M. Grilli, Phys. Rev. Lett. 94, 026401 (2005); W. Koller, A. C. Hewson, and D. M. Edwards, Phys. Rev. Lett. 95, 256401 (2005).

4 R. T. Clay and R. P. Hardikar, Phys. Rev. Lett. 95, 096401 (2005); H. Matsueda, T. Tohyama, and S. Maekawa, cond-mat/0511068 (unpublished); W. Q. Ning, H. Zhao, C. Q. Wu, and H. Q. Lin, Phys. Rev. Lett. 96, 156402 (2006); M. Tezuka, R. Arita, and H. Aoki, Phys. Rev. Lett. 95, 226401 (2005).

5 R. T. Clay and R. P. Hardikar, Phys. Rev. Lett. 95, 096401 (2005); H. Fehske, G. Wellein, G. Hager, A. Weiße, and A. R. Bishop, Phys. Rev. B 69, 165115 (2004).

6 R. T. Clay and R. P. Hardikar, Phys. Rev. Lett. 95, 096401 (2005).

7 H. Matsueda, T. Tohyama, and S. Maekawa, cond-mat/0511068 (unpublished).

8 H. Fehske, M. Holicki, and A. Weiße, in Advances in Solid State Physics, 40, 235 (2000); G. Wellein, H. Fehske, and A. P. Kampf, Phys. Rev. Lett. 81, 3956 (1998).

9 H. Fehske, G. Wellein, and A. Weiße, in Advances in Solid State Physics, 40, 235 (2000); G. Wellein, H. Fehske, and A. P. Kampf, Phys. Rev. Lett. 81, 3956 (1998).

10 H. Fehske, G. Wellein, G. Hager, A. Weiße, K. W. Becker, and A. R. Bishop, Physica B 359–361, 699 (2005).

11 L. D. Fadeev and L. A. Takhtajan, Phys. Lett. A 85, 375 (1981); M. Mohan and G. Mueller, Phys. Rev. B 27, 1776 (1983).