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Phys. Rev. B **99**, 174516 — Published 24 May 2019

DOI: [10.1103/PhysRevB.99.174516](https://doi.org/10.1103/PhysRevB.99.174516)

Pseudogap crossover in the electron-phonon system

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(Dated: April 29, 2019)

Thermodynamic properties of the square-lattice Holstein model of the electron-phonon problem with phonon frequencies small compared to the bare Fermi energy are obtained using Monte Carlo methods, a strong-coupling (bipolaronic) expansion, and a weak coupling Migdal-Eliashberg approach. Already at elevated temperatures where the charge-density wave (CDW) and superconducting (SC) correlations are very short-range, a crossover occurs as a function of increasing electron-phonon coupling, λ_0 , from a normal metallic regime to a pseudogap regime. At sufficiently low T , a SC phase is found for small λ_0 and a commensurate insulating CDW phase for large λ_0 .

Introduction – Electron-phonon interactions determine many of the electronic properties of quantum materials; this includes the normal state electrical transport properties of most metals at all but the lowest temperatures, and of course the nature of the superconducting (SC) and/or charge-density-wave (CDW) ground-states of many “conventional” materials. In all but a few cases, the dimensionless electron-phonon coupling constant, λ_0 , is of order one. Nonetheless, there is a genuine small parameter in the problem, the ratio of the phonon energy, $\hbar\omega_0$, to the bare Fermi energy, $E_F^{(0)}$. While it has been argued that Migdal-Eliashberg (ME) theory^{1,2} provides an accurate solution to this problem provided $\lambda_0 \ll E_F^{(0)}/\hbar\omega_0$, we recently showed^{3,4} that ME theory breaks down when $\lambda_0 \sim 1$, even when the nominal condition for its validity is satisfied. As was already suggested in various earlier studies,⁵ this breakdown is associated with the non-perturbative formation of bipolarons.

In the present paper, we explore the global phase diagram of the Holstein model⁶ – the paradigmatic model of the electron-phonon problem – over a broad range of temperatures, T , and λ_0 in the physically important limit $\hbar\omega_0/E_F^{(0)} \ll 1$. We have carried out extensive Monte Carlo (MC) calculations, which we then compare with the results of ME theory and with a strong-coupling expansion (in powers of $1/\lambda_0$). As shown in the schematic phase diagram in Fig 1, there are two regions separated by a crossover line, $T = T^*(\lambda_0)$; ME theory gives a good account of the physics only in the left region while a strong-coupling “polaronic” approach is accurate to the right. (Naturally, neither approach is entirely reliable close to the crossover line.)

The physics in the two regions is correspondingly distinct: In the weak coupling regime, the properties of the normal state are dominated by weakly scattered quasi-particle excitations near a well-defined Fermi surface with decay rates $\hbar\gamma \sim \lambda_0 T$ and there is a low- T superconducting ground-state with a transition temperature T_c which is proportional to $\hbar\omega_0$ times a (possibly non-monotonic) function of λ_0 . In the strong-coupling limit, there is a “pseudogap” to single-particle excitations, the normal state is a classical lattice gas of (effectively non-

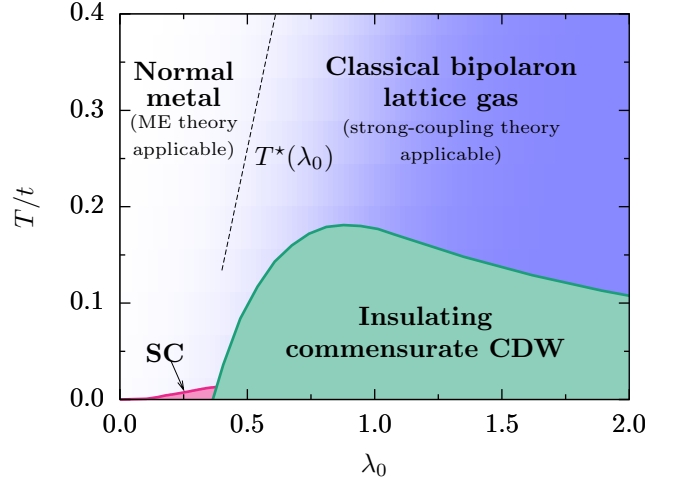


FIG. 1. Schematic phase diagram of the electron-phonon problem with small $w \equiv \hbar\omega_0/E_F^{(0)}$. Depending on details of the band-structure and the electron density, additional CDW phases can arise (including metallic ones at intermediate λ_0).

dynamical) bipolarons with binding energy $\sim \lambda_0 E_F^{(0)}$ and at low temperatures the system has a tendency to commensurate CDW states, with ordering vectors unrelated to any Fermi-surface nesting vector. Depending on the electron density there may be a sequence of transitions to higher-order commensurate states or phase separation.

We will study the Holstein Hamiltonian⁶

$$H = H_e + H_p + H_{e-p}, \quad (1)$$

$$H_e = - \sum_{ij\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} - \mu \sum_{i\sigma} n_{i\sigma}, \quad (2)$$

$$H_p = \sum_i \frac{p_i^2}{2M} + \frac{1}{2} K x_i^2, \quad (3)$$

$$H_{e-p} = \alpha \sum_{i\sigma} x_i n_{i\sigma}, \quad (4)$$

and $\omega_0 = \sqrt{K/M}$. The important dimensionless parameters are the coupling strength $\lambda_0 \equiv \alpha^2 \rho^{(0)}/K$ and the

retardation parameter $w \equiv \hbar\omega_0/E_F^{(0)}$, where $\rho^{(0)}$ is the bare ($\alpha = 0$) single-spin density of states at the Fermi energy.⁷ While there are notable exceptions, the regime of the Holstein model relevant to most materials is $w \ll 1$.

We therefore study the phase diagram and thermodynamic correlation functions of the Holstein model in the limit $w \ll 1$. In previous work,³ we carried out such a study via determinant quantum Monte Carlo (DQMC) for $w = 0.1$, but only for weak to moderate λ_0 : $0 \leq \lambda_0 \leq 0.6$. The DQMC method is challenging to employ in the strong-coupling regime due to prohibitively long autocorrelation times for temperatures much lower than $E_F^{(0)}$.⁸ Fortunately, in this strong-coupling regime especially, the results are not expected to depend strongly on M so long as $w \ll 1$, an expectation that we have confirmed where it can be tested.⁹ Thus, to analyze the full phase diagram, we will consider here the limit $M \rightarrow \infty$, corresponding to $w \rightarrow 0$. In this limit the phonons become classical variables, and the MC calculations become substantially simpler. (See the Appendix for details of the MC algorithm employed.) We have chosen parameters to avoid any non-generic band features or special commensurate densities – specifically we take the matrix t_{ij} to contain both nearest-neighbor hopping t and next-nearest-neighbor hopping t' , with the ratio $t'/t = -0.3$. We work at a fixed chemical potential, chosen such that the density is $n = 0.8$ at $T = 0.25t$. We have studied systems of linear size $L \leq 12$ with periodic boundary conditions and temperatures $T \geq t/40$.

Results – The phase diagram derived from our MC studies in the $M \rightarrow \infty$ ($w \rightarrow 0$) limit is shown in Fig. 2. For $\lambda_0 \ll 1$ we find a translationally invariant Fermi liquid ground-state. At strong-coupling $\lambda_0 \gg 1$ the low-energy degrees of freedom are bipolarons, which have a binding energy $V = \alpha^2/K$ and behave as a lattice gas of interacting hard-core classical charge $2e$ particles. (It is convenient to think of the bipolarons as hard-core bosons, but because they are non-dynamical in this limit, they in fact have no meaningful quantum statistics.) The change in the nature of the low-energy states manifests as a pseudogap in the single-particle electron spectrum, onsetting at a temperature $T^* \sim V$. At lower T and for sufficiently large λ_0 we find a $\mathbf{Q} = (\pi, \pi)$ CDW state. While ME theory is extremely accurate for $T > T^*$, we will see that it fails to describe the crossover at $T \sim T^*$, and misses the strong-coupling physics when $T < T^*$ entirely. By contrast, the strong-coupling expansion (also discussed below) gives a satisfactory account of the system in the strong-coupling regime; in particular, the CDW phase boundary labeled $T_c^{\text{Ising}} \sim 1/\lambda_0$ in the figure was computed to leading order in the strong-coupling expansion for the same parameters as in the MC study. Variations of the density with temperature and coupling strength are shown in Figure 3.

We have previously carried out DQMC calculations for this model with large but finite M such that $w = 0.1$. We expect the thermodynamic properties for $w = 0$ to be similar to those with $w = 0.1$ when $T > \hbar\omega_0$ or to

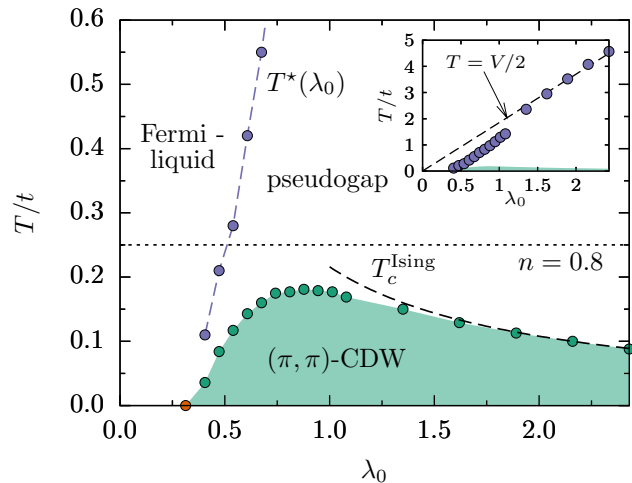


FIG. 2. Phase diagram of the Holstein model in the $M \rightarrow \infty$ limit with $t'/t = -0.3$. Chemical potential has been chosen such that $n = 0.8$ for $T = 0.25t$. The inset shows the phase diagram extended to higher temperatures where, for $\lambda_0 \gtrsim 1$, T^* becomes equal to half the bipolaron binding energy, $V/2$, as expected from the strong-coupling limit. See text for additional details.

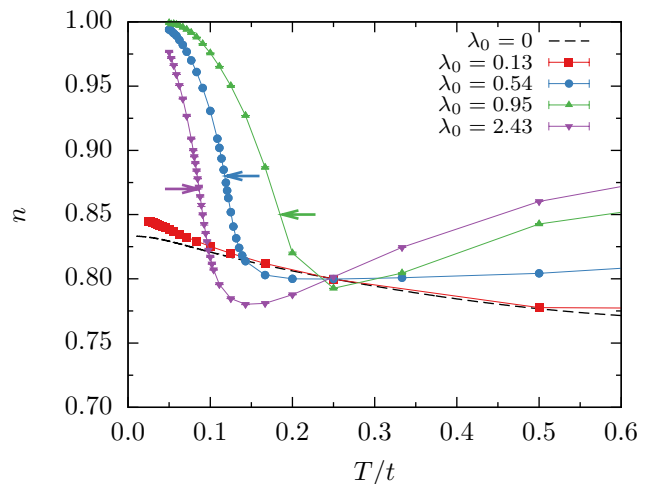


FIG. 3. Density n as a function of T for various coupling strengths. The dashed line shows the evolution of the density for non-interacting electrons. Arrows indicate T_{cdw} inferred from finite-size scaling. Linear system size is $L = 12$.

the right of the $T^*(\lambda_0)$ line. While we have not explicitly tested this in all cases (especially at large λ_0 and low T , where the $w \neq 0$ DQMC is most difficult), we have verified the validity of this expectation wherever we have $w \neq 0$ results. Even to the left of the $T^*(\lambda_0)$ line, the results with $w = 0$ and $w = 0.1$ differ little down to temperatures that are a small fraction of $\hbar\omega_0$. However, in this weak coupling regime, for finite w (but still $w \ll 1$) we expect a SC transition at $T_c \sim \hbar\omega_0 \exp(-1/\lambda)$. This accounts for the one qualitative difference between Fig.

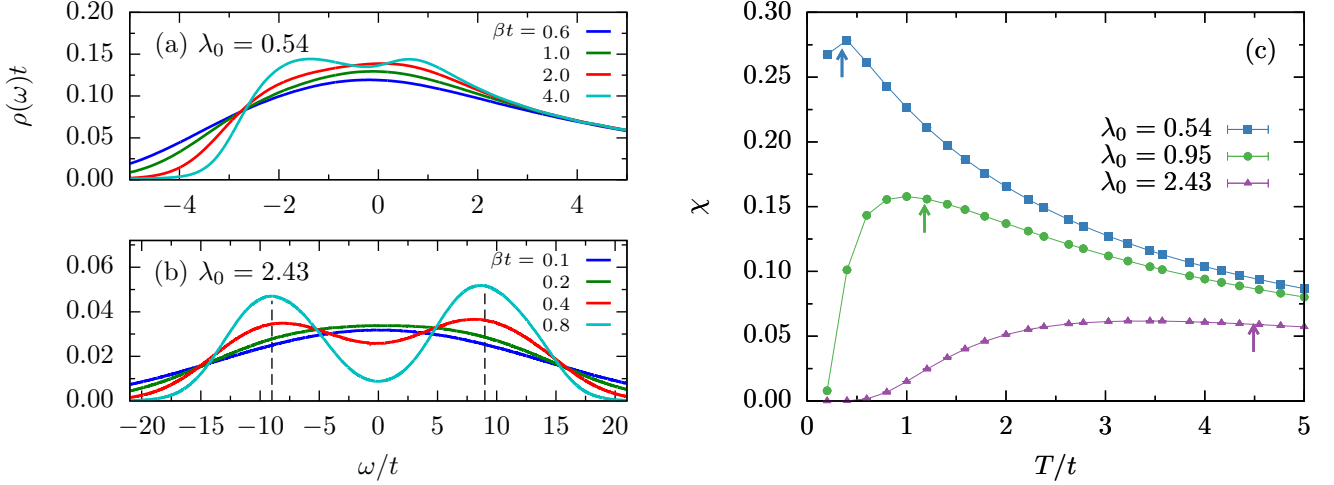


FIG. 4. DOS as a function of energy relative to E_F at various T (a) for $\lambda_0 = 0.54$ and (b) for $\lambda_0 = 2.43$. The dashed vertical lines indicate the bipolaron binding energy $\pm V$. Linear system size is $L = 12$. Delta functions are broadened with $\eta \approx 10/L^2$. T^* is identified as the highest temperature at which there is a local minimum at $\omega = 0$. (c) The spin susceptibility as a function of T for various λ_0 ; the arrows indicated T^* inferred from the DOS. Linear system size is $L = 10$.

1 and 2; in the former we have added a SC phase below a critical temperature computed according to ME theory,³ while of course $T_c = 0$ in the $M \rightarrow \infty$ limit.¹⁰

CDW phase – At zero temperature, mean-field theory is exact in the $M \rightarrow \infty$ limit and gives a first order transition to a $\mathbf{Q} = (\pi, \pi)$ CDW state at a critical coupling strength $\lambda_0 = \bar{\lambda}_0 \approx 0.31$, indicated by the orange circle in Fig. 2. At fixed density, the system would phase separate into a region with $n = 1$ and a region with $n < 0.8$. For $\lambda_0 > \bar{\lambda}_0$, the (π, π) state persists to $T > 0$ and the finite temperature transitions we have observed appear continuous. (Presumably, this is not the case at low enough T since, on theoretical grounds, if there is a first order transition at $T = 0$ one would expect it to persist to small non-zero T .) The CDW phase boundary is identified by finite-size scaling of the phonon correlation function at wave-vector \mathbf{Q} ,

$$D(\mathbf{Q}) = \frac{1}{L^2} \sum_{ij} e^{i\mathbf{Q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} \langle x_i x_j \rangle, \quad (5)$$

where L is the linear system size, assuming the transition to be in the Ising universality class. Details of this analysis are provided in the Appendix.

From a strong-coupling expansion in $1/\lambda_0$ one finds the effective Hamiltonian for the system (which gives a valid description for temperatures $T \ll V$) is an antiferromagnetic Ising model in a uniform external field:^{11,12}

$$H_{\text{eff}} = \sum_{ij} J_{ij} \tau_i \tau_j - h \sum_i \tau_i, \quad (6)$$

where τ_i are classical Ising variables taking on the values ± 1 and $J_{ij} = 2t_{ij}^2/U$. The relation to the electronic degrees of freedom is that $\tau_i = 1$ if a site is occupied by a bipolaron and zero otherwise. The density n of the original electrons and the magnetization

m of the Ising spins are related by $n = 1 + m$. In the parameter regime where the nearest-neighbor J is much stronger than all further neighbor couplings and also near to half-filling ($m = 0$), this model has a transition from a paramagnetic phase to a (π, π) antiferromagnetic phase at a temperature $T_c^{\text{Ising}} \sim J$. Depending on m and the nature of the further neighbor couplings there may be additional ordering transitions or phase separation at lower temperatures.¹³ We have computed T_c^{Ising} for the parameters relevant to the model under consideration – nearest-neighbor $J = 2t^2/U$, next-nearest-neighbor $J' = 2t'^2/U$, and external field h tuned such that $m(T = 0.25t) = -0.2$ – and in Figure 2 we show that the transition temperature coincides very accurately with T_{cdw} of the full Holstein model for $\lambda_0 \gtrsim 1$.

Pseudogap – The pseudogap region can be delimited by various crossover temperature scales. We define $T^*(\lambda_0)$ as the temperature below which the electronic density of states (DOS) $\rho(\omega)$ develops a minimum at $\omega = 0$, similar to the conventional definition of the pseudogap temperature in various correlated materials. The DOS for a given phonon configuration X is

$$\rho_X(\omega) = \frac{1}{L^2} \sum_{\lambda} \delta(\omega - E_{\lambda}[X]), \quad (7)$$

where E_{λ} are the single-particle energies in the phonon configuration X . The DOS is then obtained by averaging over phonon configurations (This procedure is explained in more detail in the Appendix). In practice the delta functions in (7) are resolved with a Lorentzian broadening, with broadening parameter η chosen to be on the order of the finite size gaps in the single-particle spectrum for a given system size. The DOS for representative weak and strong-coupling values is shown in Figure 4.

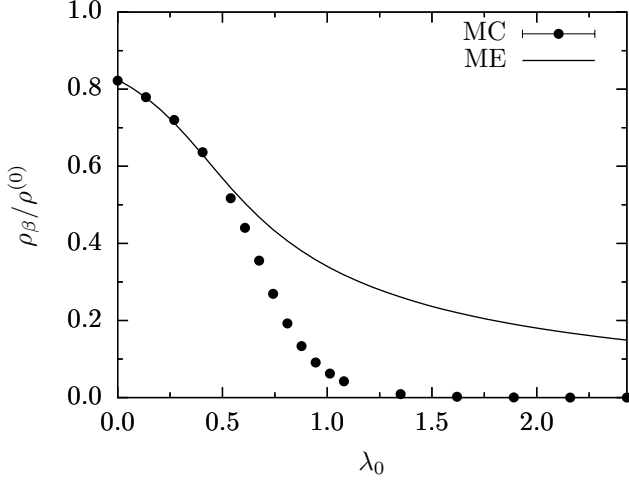


FIG. 5. Temperature averaged DOS at the Fermi-energy ρ_β at $T = 0.25t$, normalized by the zero-temperature, non-interacting DOS $\rho^{(0)}$. At this elevated temperature $\rho_\beta < \rho^{(0)}$ even for $\lambda_0 = 0$. The breakdown of ME theory occurs for $\lambda_0 \approx 0.5$. Linear system size is $L = 12$.

The appearance of a pseudogap is also evident in thermodynamic observables. In the $w = 0$ limit of the current model, the charge and spin susceptibilities are degenerate and we therefore define a single static susceptibility

$$\chi = \frac{\beta}{L^2} \left(\langle \hat{N}^2 \rangle - \langle \hat{N} \rangle^2 \right) = \frac{\beta}{L^2} \left(\langle \hat{M}^2 \rangle - \langle \hat{M} \rangle^2 \right), \quad (8)$$

where $\hat{N} = \sum_i \hat{n}_i$ and $\hat{M} = \sum_i \hat{S}_i^z$. In Figure 4 we see there is indeed a depression of χ below T^* .

ME theory – Migdal-Eliashberg (ME) theory purports to solve the electron-phonon problem for any coupling strength λ_0 , provided the product $\lambda_0 w \ll 1$.^{1,2} For $w = 0$ the ME theory should therefore be valid for arbitrary λ_0 . To assess the validity of this statement we compare the single-particle DOS computed within ME theory to that obtained with MC results. Our ME calculations are carried out in imaginary time and therefore comparison with dynamical quantities (e.g., single-particle DOS) requires analytic continuation. Rather than dealing with complications associated with analytic continuation we will work with a proxy for the low-energy DOS:

$$\rho_\beta \equiv \frac{\beta}{\pi} G(\mathbf{x} = 0, \tau = \beta/2) = \frac{\beta}{2\pi} \int d\omega \frac{\rho(\omega)}{\cos(\beta\omega/2)}. \quad (9)$$

This quantity is essentially the single-particle DOS averaged over an energy window of order the temperature. At low-temperatures $\rho(E_F) \approx \rho_\beta$. In Figure 5 we show ρ_β computed with MC and within ME theory, at $T = 0.25t$ where the density in both calculations is $n = 0.8$. We find ME becomes qualitatively incorrect for $\lambda_0 \gtrsim 0.5$, where the MC shows a precipitous drop due to the onset of the pseudogap ($T^* \approx 0.25t$ for $\lambda_0 = 0.5$), while the ME shows a much weaker dependence. We emphasize

that this temperature is well above T_{cdw} ($T_{\text{cdw}} \approx 0.1t$ for $\lambda_0 = 0.5$) and therefore the breakdown of ME theory is unrelated with the onset of CDW order. Indeed, we find that T_{cdw} drops rapidly as the density decreases from $n = 1$, while T^* is essentially unchanged. Rather, the breakdown occurs because of a dramatic rearrangement in the low-energy spectrum upon entering the pseudogap regime. When $\lambda_0 \gtrsim 1$, and even when $\lambda_0 w = 0$, the ME perturbation theory breaks down because it is a perturbative expansion around the wrong state.

Conclusion – While ME theory works well for sufficiently weak coupling, it breaks down to the right of the $T^*(\lambda_0)$ line where the system is described by a classical lattice gas whose low-energy excitations are bipolarons. Because T^* exceeds significantly the ordering temperature T_{cdw} we do not associate the breakdown of ME theory with a competing order or fluctuations near T_{cdw} . Furthermore, we emphasize that the CDW transition is unrelated to features of the Fermi surface but is associated with the commensurate ordering expected from the Ising description of the system in strong coupling.

Interesting materials typically have multiple phonon branches, often multiple electronic bands crossing the Fermi energy, and generally more structured electron-phonon coupling, so quantitative comparison with the results for the Holstein model are of course not possible. However, we feel that aspects of the present results are of general relevance. Two aspects of the results, in particular, are relevant to phonon-mediated superconductivity. On the one hand, the breakdown of ME theory when $\lambda_0 \sim 1$ appears to be unavoidable; for example, seeking ways to prevent a lattice instability (e.g. CDW ordering) does *not*, by itself, extend the range of validity of ME theory. Moreover, while large λ_0 can indeed produce a large pairing scale, the resulting bipolaron formation is accompanied by a drop in the superconducting susceptibility. These results further corroborate our earlier inference⁴ that there is an optimal value of $\lambda_0 \sim 1$ at which T_c is maximal, and that T_c always drops quickly to zero for larger λ_0 .

In exploring whether the optimal λ_0 obtained here is consistent with experimental data, it is important to distinguish the bare value of the electron-phonon coupling – our λ_0 – from the renormalized value, λ , which can be extracted, for example, from tunneling data. The induced phonon softening that is prominent at larger coupling strength results in values of $\lambda > \lambda_0$. For example, for the Holstein model with the same parameters studied here,³ $\lambda_0 = 0.5$ corresponds to $\lambda \approx 2$. In this context, we note that in the famous Allen-Dynes¹⁴ compilation of experimental values of λ and T_c for a large number of conventional superconductors, all the entries are roughly in the range $\lambda \lesssim 2$. The presence of an apparent upper bound on λ in SCs is something that is not expected on the basis of ME theory.

Acknowledgements: We would like to acknowledge helpful discussions with Yoni Schattner and Edwin Huang. SAK and IE were supported, in part, by NSF grant #

DMR-1608055 at Stanford. DJS was supported by the Scientific Discovery through Advanced Computing (SciDAC) program funded by U.S. Department of Energy, Office of Science, Advanced Scientific Computing Re-

search and Basic Energy Sciences, Division of Materials Sciences and Engineering. Computational work was performed on the Sherlock computing cluster at Stanford University.

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- ¹ A. Migdal, Sov. Phys. JETP **7**, 996 (1958).
² G. Eliashberg, Sov. Phys. JETP **11**, 696 (1960).
³ I. Esterlis, B. Nosarzewski, E. W. Huang, B. Moritz, T. P. Devereaux, D. J. Scalapino, and S. A. Kivelson, Phys. Rev. B **97**, 140501 (2018).
⁴ I. Esterlis, S. A. Kivelson, and D. J. Scalapino, npj Quantum Materials **3**, 59 (2018).
⁵ The breakdown of ME theory has been discussed previously from the perspective of dynamical mean field theory^{11,15–18} and through an analysis¹⁹ of ground-state properties in the $M \rightarrow \infty$ limit.
⁶ T. Holstein, Annals of Physics **8**, 325 (1959).
⁷ Note that beyond the weak coupling regime, $\lambda_0 \ll 1$, there is a non-trivial relation³ between the bare quantities and the “renormalized” or “physical” values of λ , $\rho(E_F)$, and the phonon dispersion, $\omega_{\mathbf{q}}$.
⁸ M. Hohenadler and T. C. Lang, in *Computational Many-Particle Physics* (Springer, 2008) pp. 357–366.
⁹ In Ref. 20, a similar comparison was made between results with $w = 0$ and non-zero w for the half-filled Holstein model.
¹⁰ This also means we cannot directly address the SC to CDW transition in our calculations. However, from the nature of the zero temperature phase diagram in the $\omega_0 = 0$ limit (discussed in the Appendix), it is probable that there is a sequence of transitions, $SC \rightarrow SC+CDW \rightarrow CDW$, as a function of increasing coupling λ_0 for a small but non-zero phonon frequency. The first of these transitions from $SC \rightarrow SC+CDW$ is likely to be first order.
¹¹ J. K. Freericks, M. Jarrell, and D. J. Scalapino, Phys. Rev. B **48**, 6302 (1993).
¹² E. W. Carlson, V. J. Emery, S. A. Kivelson, and D. Orgad, “Concepts in high temperature superconductivity,” in *Superconductivity: Conventional and Unconventional Superconductors*, edited by K. H. Bennemann and J. B. Ketterson (Springer Berlin Heidelberg, Berlin, Heidelberg, 2008) pp. 1225–1348.
¹³ K. Binder and D. P. Landau, Phys. Rev. B **21**, 1941 (1980).
¹⁴ P. B. Allen and R. C. Dynes, Phys. Rev. B **12**, 905 (1975).
¹⁵ A. J. Millis, R. Mueller, and B. I. Shraiman, Phys. Rev. B **54**, 5389 (1996).
¹⁶ P. Benedetti and R. Zeyher, Phys. Rev. B **58**, 14320 (1998).
¹⁷ D. Meyer, A. C. Hewson, and R. Bulla, Phys. Rev. Lett. **89**, 196401 (2002).
¹⁸ M. Capone and S. Ciuchi, Phys. Rev. Lett. **91**, 186405 (2003).
¹⁹ A. S. Alexandrov, EPL (Europhysics Letters) **56**, 92 (2001).
²⁰ M. Weber and M. Hohenadler, Phys. Rev. B **98**, 085405 (2018).