Bipolaron liquids at strong Peierls electron-phonon couplings

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We use the density matrix renormalization group method to study a one-dimensional chain with Peierls electron-phonon coupling, which describes the modulation of the electron hopping by lattice distortions. We demonstrate that this system is stable against phase separation in the dilute density limit. We only find phase separation numerically for large couplings for which the linear approximation for the electron-phonon coupling becomes invalid; this behavior can be stabilized in a narrow sliver of the physical parameter space if the dispersion of the phonons is carefully tuned. These results indicate that in the dilute electron density limit, Peierls bipolaron liquids are generically stable, unlike in other models of electron-phonon coupling. We show that this behavior extends to finite carrier concentrations of up to quarter filling. This stability of low-density, light-mass bipolaron liquids in the Peierls model opens a path to high-T_c superconductivity based on a bipolaronic mechanism, in higher dimensions.

Introduction. A primary goal in condensed matter physics targets the discovery and understanding of unusual phases of matter that arise from strong correlations. Correlated electron-lattice systems manifest in fascinating ways in many experimentally relevant situations, such as polaronic phenomena in the dilute electron density limit [1–5], and charge order and superconductivity at finite electron concentrations [6–8]. In particular, the search for alternative microscopic mechanisms for high-T_c superconductivity has stimulated a renewed interest in the study of bipolarons—bound states of two polarons, where a polaron is an electron dressed by a cloud of phonons—in the presence of different forms of electron-lattice coupling.

Recent work [4] has in fact demonstrated that the Peierls electron-phonon coupling [9–11], which describes the modulation of the electron hopping due to lattice distortions, gives rise to strongly bound but light-mass bipolarons. As a result, these should remain phase coherent up to high temperatures, opening a possible route to phonon-mediated high-T_c superconductivity whose fingerprints may have already been observed experimentally in the material $Ba_{1-x}K_xBiO_3$ [12,13]. A possible hindrance to this scenario are competing instabilities that could favor a different order. For example, for the more studied generalized Holstein [14,15] and Fröhlich [16,17] models in which the lattice distortion modulates the electron's on-site energy, phase separation sets in at fairly weak electron-phonon couplings for low carrier concentrations [18–20]. This is because bipolarons

in these models become increasingly heavy at stronger couplings [21–23] and experience a phonon-mediated long-range electron-electron attraction. The latter favors clustering of bipolarons in order to minimize the total energy, thus inducing phase separation. This phenomenon presents a major obstacle to bipolaronic superconductivity (at any temperature) in these models. In contrast, as stated earlier, the Peierls coupling favors light bipolarons even at very large couplings because it mediates effective *pair-hopping* interactions that enhance the kinetic energy of bipolarons [4]. As such, it is unclear whether phase separation occurs in the Peierls model at finite electronic densities. In this regime, the lack of controllable analytical approaches calls for the use of unbiased numerical techniques.

In this Letter, we use the density matrix renormalization group (DMRG) method [24] to show that in one dimension (1D), a dilute liquid of bipolarons is stable against phase separation up to strong Peierls electron-phonon couplings. Phase separation occurs only when lattice distortions are so large such that the linear approximation breaks down and the model becomes unphysical. Refinements such as the inclusion of a finite phonon dispersion may render phase separation possible for sufficiently large couplings, but also pushes the critical coupling to larger values. We illustrate this main result in Fig. 1 where we contrast the stability of a liquid of Peierls bipolarons with the behavior of a generalized extended Holstein model in which phase separation sets in at very low couplings. We also confirm that this phenomenology extends to finite electron concentrations up to quarter filling (see Fig. 4).

Instability of polarons or bipolarons to phase separation operates more effectively in lower dimensions. Therefore, given our results in 1D, we expect the stability of a liquid of

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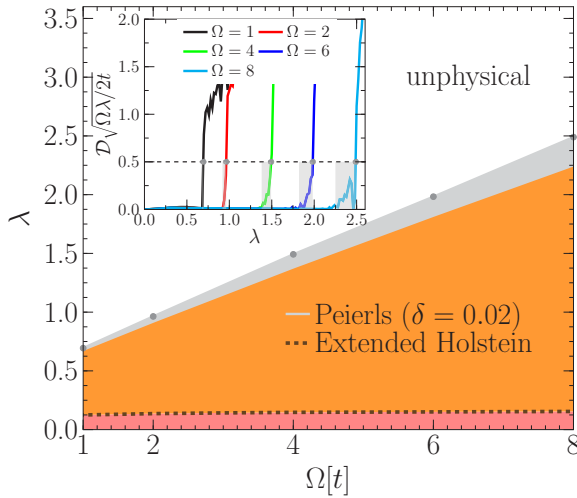†alberto.nocera@ubc.ca
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FIG. 1. Main panel: Phase diagram of the 1D Peierls model contrasted against that of the extended Holstein (EH) model in the dilute electron density limit. The latter represents a lattice model that mimics the physics of the Fröhlich coupling, see Ref. [18], for which $\lambda = g^2/(2\Omega t)$. The phase boundary defined by $\lambda_c(\Omega)$ separates a stable liquid of bipolarons from phase separation. For the Peierls model, the bipolaron liquid is stable for $\lambda < \lambda_c$ (orange region), becomes unstable to phase separation for $\lambda_c < \lambda < \lambda_s$ (grey region), and the latter quickly gives way to an unphysical regime for $\lambda > \lambda_s$ (white region), see text for details. For the EH model, the dashed line labels critical values $\lambda_c(\Omega)$ separating a stable liquid of bipolarons for $\lambda < \lambda_c$ (red-orange region) from the phase separation for $\lambda > \lambda_c$ (all regions above). In clear contrast to the EH model, in the Peierls model the bipolaron liquid is stable up to much larger λ, with $\lambda_c \to \infty$ as $\Omega \to \infty$. These DMRG results are for a system with $L = 32$ sites, $N = 6$ electrons and using a finite dispersion parameter $\delta = 0.02$. Inset: Staggered displacement amplitude $\mathcal{D}$, defined in Eq. (1), as a function of λ for different values of Ω in the Peierls model. The horizontal dashed line at $1/2$ defines the limit beyond which phonon displacements become unphysical.

Peierls bipolarons to extend to higher dimensions. This, coupled with the arguments of Ref. [4] showing that the Peierls model realizes light-mass bipolarons, further supports the possibility of high-T_c superconductivity based on a bipolaronic mechanism in physically relevant dimensions [25,26].

Model. We consider the one-dimensional Peierls model of electron-phonon coupling [9–11,27,28]

$$\mathcal{H} = \mathcal{H}_e + \mathcal{H}_{ph} + V_{e-ph}.$$

Here

$$\mathcal{H}_e = -t \sum_{i,\sigma} c_{i,\sigma}^\dagger c_{i+1,\sigma} + \text{H.c.}$$

describes nearest-neighbor hopping of electrons of spin $\sigma \in \{\uparrow, \downarrow\}$ in a single electronic band with creation operator $c_{i,\sigma}^\dagger$ at site $i \in \{1, .., L\}$, and number operator $\hat{n}_i = \sum_\sigma \hat{n}_{i,\sigma}$. The Peierls electron-phonon coupling describes the modulation of the hopping integral due to lattice distortions in the linear approximation:

$$V_{e-ph} = g \sum_{i,\sigma} (c_{i,\sigma}^\dagger c_{i+1,\sigma} + \text{H.c.})(b_i^\dagger + b_i - b_{i+1}^\dagger - b_{i+1}).$$

Phonons belong to an optical Einstein mode of frequency Ω (we set $\hbar = 1$) with

$$\mathcal{H}_{ph} = \Omega \sum_i b_i^\dagger b_i + \delta \frac{\Omega}{4} \sum_i (b_i^\dagger + b_i - b_{i+1}^\dagger - b_{i+1})^2.$$

Here, a phonon is described by a boson creation operator $b_i^\dagger$ at site $i \in \{1, .., L\}$. The δ term gives a dispersion to the optical phonons (we set the lattice spacing $a = 1$ and oscillator mass $M = 1$): $\Omega_q = \Omega\sqrt{1 + 4\delta \sin^2(q/2)}$ shows hardening at the zone boundary $q = \pm\pi$ if $\delta \neq 0$. We characterize the strength of Peierls electron-phonon coupling via the dimensionless coupling $\lambda = 2g^2/(\Omega t)$.

Methods. We study $N \leqslant 16$ electrons in the zero magnetization sector $S_{\text{Tot}}^z = 0$ of the Peierls model on chains of lengths $L \leqslant 48$. We use DMRG to compute the ground state, utilizing up to $n_{\text{ph}_{\text{max}}} + 1 = 20$ phonon states to represent the local phonon Hilbert space. In the literature, DMRG has been often used to study one dimensional correlated Hamiltonians with Holstein electron-phonon coupling, seldomly with Peierls electron-phonon coupling [29–43]. Our numerical results were converged with respect to the bond dimension m. A maximum $m = 600$ provides convergence with a truncation error smaller than 5×10^{-7} for open boundary conditions (OBC) and 5×10^{-6} for periodic boundary conditions (PBC), see the Supplemental Materials for more information [44].

Note that unlike in quantum Monte Carlo approaches, where an explicit cutoff on the amplitude of the lattice distortions is introduced to avoid unphysical changes in the sign of the hopping term [45,46] or a restricted interval of interaction strengths is explored, in DMRG simulations one may use the δ phonon term as a Lagrange multiplier that energetically penalizes unphysical changes in the sign of the hopping, i.e., as a physical constraint on the length of the bonds.

Dilute electron density limit. Previous work showed that Peierls bipolarons are stable against dissociation into single polarons for all λ, unless an extremely large Hubbard repulsion is present (a possibility ignored in this paper) [4]. We first analyze the stability of a dilute liquid of these Peierls bipolarons. We use DMRG to find the ground-state (GS) energies E_N for $N = 1, 2, 4, 6$ electrons on a chain with $L = 32$ sites shown in Fig. 2. We define $\Delta_2 = 2E_1 - E_2$, $\Delta_4 = 2E_2 - E_4$, $\Delta_6 = E_2 + E_4 - E_6$ and study their dependence on λ in Fig. 2(a). In the thermodynamic limit, all $\Delta_N \geqslant 0$: $\Delta_N = 0$ indicates that smaller complexes, each composed of fewer than N particles, are energetically favorable (e.g., $\Delta_4 = 0$ means that the $N = 4$ GS consists of two bipolarons), while $\Delta_N > 0$ implies that bound state of the N carriers is more stable. Figure 2(a) shows that $\Delta_2 > 0$ for all λ, confirming that the $N = 2$ GS always corresponds to a bipolaron, in agreement with [4]. Both Δ_4 and Δ_6 only become positive above roughly the same $\lambda_c \gtrsim 0.8$, showing the tendency of all carriers present in the system to coalesce if $\lambda \geqslant \lambda_c$. [We note that negative Δ_2, Δ_4 values for $\lambda < \lambda_c$ are due to finite-size effects, see the inset of Fig. 2(a).] (Additional analysis based on Maxwell construction is presented in the Supplemental Material [44]). Note that below λ_c we find $E_{2N} = NE_2$, and E_2 agrees with the single bipolaron energy of Ref. [4]. This confirms that this dilute bipolaron liquid is formed of essentially isolated bipolarons whose properties follow from
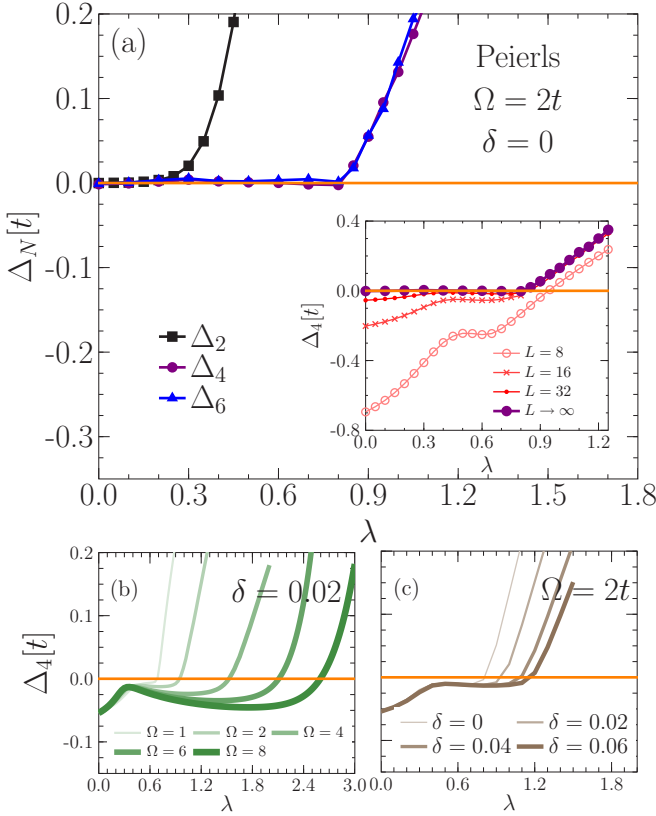
FIG. 2. (a) Main panel: Energies $\Delta_2 = 2E_1 - E_2$, $\Delta_4 = 2E_2 - E_4$, $\Delta_6 = E_2 + E_4 - E_6$ against λ for $\Omega = 2t$, where E_N is the GS energy for N electrons on a chain of infinite length. For $\lambda > \lambda_c \approx 0.8$, $\Delta_4 > 0$ and $\Delta_6 > 0$ signal that the $N = 4, 6$ carriers bind, indicating phase separation (see text for more details). Inset: Scaling of Δ_4 with system size L proves that for $\lambda < \lambda_c \approx 0.8$, $\Delta_4 \to 0$ as $L \to \infty$. The scaling was performed using data up to $L = 64$ (not shown) and assuming $\Delta_4(L) = a_0 + \frac{a_2}{L^2} + \frac{a_4}{L^4}$ for $\lambda < \lambda_c \approx 0.8$ and $\Delta_4(L) = a_0 + \frac{a_2}{L}$ for $\lambda > \lambda_c \approx 0.8$. (b) Δ_4 as a function of λ for various values of the phonon frequency Ω, at $\delta = 0.02$. (c) Δ_4 as a function of λ for various values of δ of the phonon dispersion, at $\Omega = 2t$.

Ref. [4]; in other words they continue to be of light-mass and strongly bound in the dilute liquid phase.

Figure 2(b) shows the evolution of λ_c with Ω, which we use to identify the location of the phase boundary between the bipolaron liquid and phase separation in the phase diagram of Fig. 1. Figure 2(c) shows that λ_c drifts to larger values with increasing δ, indicating that the instability of bipolarons to clumping (phase separation) is unlikely in realistic systems (see Supplemental Material [44] for an extended discussion). This drift occurs due to a competition before the onset of phase separation between the δ term, which disfavors dimerization, and the electron-phonon interaction, which drives dimerization at large couplings. Special combinations of δ and λ render phase separation possible within the domain of physicality of the model, as we discuss next.

In the linear approximation, $t(x_i - x_j) \approx t - g\sqrt{2\Omega}(x_i - x_j)$, where $x_i \equiv \sqrt{\frac{1}{2\Omega}}(b_i^\dagger + b_i)$. This approximation is only valid for small distortions $\langle x_i - x_j \rangle \ll t/g\sqrt{2\Omega}$. As we show
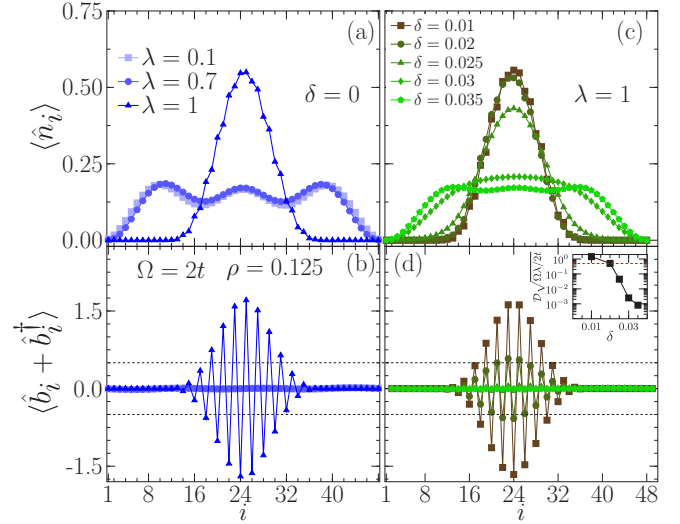

FIG. 3. GS expectation values of the average occupation number $\langle \hat{n}_i \rangle$ [(a),(c)], lattice distortion $\langle b_i + b_i^\dagger \rangle$ [(b),(d)]. The results were obtained for a chain with $L = 48$ and OBC and with $N = 6$ electrons ($\rho = 0.125$) and at phonon frequency $\Omega = 2t$. Panels (a) and (b) show results for various λ for dispersionless phonons $\delta = 0$. Panels (c) and (d) show results for various δ at strong coupling $\lambda = 1$ corresponding to the unphysical regime. Inset of (d) shows the staggered displacement amplitude [Eq. (1)] as a function of δ; the horizontal red dashed line at $1/2$ sets the limit of physicality for the phonon displacements.

next, we always find evidence for lattice dimerization at the onset of phase separation: $\langle x_i \rangle \sim (-1)^i \langle x \rangle$. Thus, here the model remains physical iff $|\langle b_i^\dagger + b_i \rangle| \ll \frac{t}{2g} = \frac{1}{2}\sqrt{\frac{2t}{\Omega\lambda}}$. We define the *staggered* dimerization amplitude

$$\mathcal{D} = \frac{1}{S}\sum_{j=1}^{S}|\langle b_j^\dagger + b_j \rangle|, \tag{1}$$

computed over the *typical* size $S \equiv S(\rho, \Omega, \lambda)$ of the dimerized region characterized by finite displacements (width at half-maximum of the displacement profile) as a proxy for the viability of the linear model: results are physical only when $\mathcal{D}\sqrt{\frac{\Omega\lambda}{2t}} < \frac{1}{2}$. The inset of Fig. 1 shows that this quantity is very small at small λ, but increases very sharply at larger λ. $\mathcal{D}\sqrt{\frac{\Omega\lambda}{2t}}$ crosses the physicality limit of $1/2$ (gray dots in inset) marking the limits in the phase diagram (gray dots in main figure) beyond which the model become definitively unphysical due to the breakdown of the linear approximation. This means phase separation exists only within a narrow sliver of the phase diagram whose width would be further reduced if a more stringent criterion $\mathcal{D}\sqrt{\frac{\Omega\lambda}{2t}} \ll \frac{1}{2}$ is used. Unraveling the physics at very large electron-phonon couplings requires inclusion of nonlinear corrections to the coupling in the model, which is beyond the scope of this work.

Figure 3 presents evidence that coalescing of bipolarons at $\lambda > \lambda_c$ represents a signature of true phase separation. Figures 3(a)–3(c) show the average electron density $\langle \hat{n}_i \rangle$ as a function of position i for a chain with OBC and $L = 48$
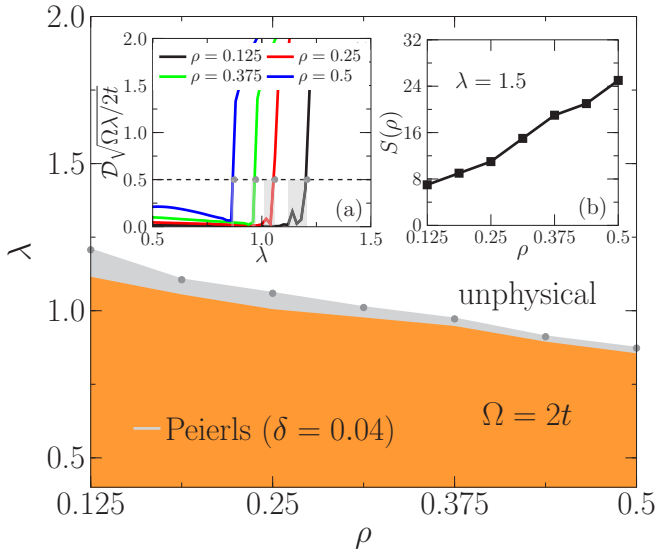
FIG. 4. Main panel: Phase diagram of the 1D Peierls model as a function of the carrier filling $\rho = N/L$ ($\rho = 0.5$ corresponds to quarter filling) and λ from DMRG for a system with $L = 32$ sites and $\Omega = 2t$. Inset (a): Staggered displacement amplitude [Eq. (1)] as a function of λ for various ρ. The horizontal dashed line at $1/2$ sets the limit of physicality for the phonon displacements. Inset (b): Size of the dimerized region in the phase separated phase as a function of ρ.

sites and $N = 6$ electrons (nominal density $\rho = 0.125$) and $\Omega = 2t$. In the $\delta = 0$ limit, depicted in Fig. 3(a), we observe for $\lambda < \lambda_c$ a density profile with $N/2$ peaks characteristic of a liquid of bipolarons; as λ grows and crosses λ_c, the electrons clump into an electron-rich region forming a Gaussian-like droplet with a maximum density $n = 0.5$, surrounded by electron-free regions, a behavior characteristic of phase separation [47]. We emphasize that due to the use of OBC, the electron-rich, phase-separated region is centered at $L/2$. Simulations with PBC (see Supplemental Material [44]) give identical results at strong coupling, but with the center of the electron-rich region not pinned to a specific site. Figure 3(c) establishes that even at strong coupling, e.g., $\lambda = 1$, a sufficiently large δ melts the electron-rich region re-instating the bipolaronic liquid phase. Figures 3(b)–3(d) demonstrates that the lattice indeed dimerizes: $(-1)^i \langle b_i + b_i^\dagger \rangle > 0$ in the core of the electron-rich region; for $\lambda > \lambda_c$. In particular, Fig. 3(b) shows that for $\lambda = 1 > \lambda_c$ the phonon displacement amplitude within the electron-rich region far exceeds the physically allowed limit $\mathcal{D}\sqrt{\Omega\lambda/2t} = 1/2$. Figure 3(d) shows that δ can be *fine tuned* so that an electron-rich region is accompanied by lattice dimerization within the stability limits of the model. We have also verified (not shown) that double occupancy in the core of the electron-rich region is very small $\langle \hat{n}_{i\uparrow}\hat{n}_{i\downarrow} \rangle \simeq 0.125$, which implies that a moderate repulsive Hubbard U will not affect these results significantly, see, e.g., [48].

Small but finite electron concentrations. Having established the stability of bipolarons in the extremely dilute limit, we also confirm the stability of the bipolaron liquid for finite densities $\rho = 0.125 - 0.5$. We construct the phase diagram as a function of ρ in Fig. 4 for $\delta = 0.04$. We find, as before, a robust bipolaronic liquid that is stable up to very large values of λ, almost everywhere the linear approximation is valid. The critical value λ_c above which phase separation is energetically favorable decreases with electronic filling. In the Supplemental Material [44], we determine the critical δ at strong coupling $\lambda = 1$ such that the dimerized phonon displacement amplitude falls within the physical limit, finding that δ_c increases nearly linearly from the dilute density limit up to quarter filling. These results conclusively demonstrate the robustness of the bipolaron liquid phase in the Peierls model *beyond* the extremely dilute density limit. This is particularly relevant for commensurate densities such as quarter filling $\rho = 0.5$, where instabilities to orders other than phase separation might have been anticipated.

Conclusions. We have numerically studied a model with linear Peierls electron-phonon coupling on a 1D chain, and proved that a dilute bipolaron liquid is stable against phase separation up to large couplings $\lambda < \lambda_c$. For $\lambda > \lambda_c$ we present evidence for a new type of phase separation, with several interesting properties including lattice dimerization within the electron-rich region surrounded by electron poor, undimerized regions. The region of validity of the linear approximation terminates close to λ_c, so that whether phase separation occurs in the model upon inclusion of higher-order, nonlinear couplings, remains an open question. Furthermore, we have confirmed the stability of bipolaron liquids in most of the physical parameter space for small carrier densities extending up to quarter filling. In contrast, in more studied models of electron-phonon coupling like in the extended Holstein model, phase separation sets in at a much smaller λ_c. These results reinforce the possibility of bipolaronic high-temperature superconductivity in models with Peierls-like coupling at low carrier densities, a scenario believed to be impossible for other forms of electron-phonon coupling [49].

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