Resonating valence bond states and other surprises in an electron-phonon system

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We study a simple electron-phonon model on square and triangular versions of the Lieb-lattice using an asymptotically exact strong coupling analysis. At zero temperature and electron density $n = 1$ (one electron per unit cell), for various ranges of parameters in the model, we exploit a mapping to the quantum dimer model to establish the existence of a spin-liquid phase with $\mathbb{Z}_2$ topological order (on the triangular lattice) and a multi-critical line corresponding to a quantum critical spin liquid (on the square lattice). In the remaining part of the phase diagram, we find a host of charge-density-wave phases (e.g. valence-bond crystals), a conventional $s$-wave superconducting phase, and with the addition of a small Hubbard $U$ to tip the balance, a phonon-induced $d$-wave superconducting phase. Under a special condition, we find a hidden pseudo-spin $SU(2)$ symmetry that implies an exact constraint on the superconducting order parameters.

The electron-phonon interaction plays an essential role in the physics of quantum materials, e.g. for Bardeen-Cooper-Schrieffer superconductivity (SC) and typical charge or bond-density-wave ordering [1–3]. In the past few years, it has become increasingly clear that electron-phonon interactions can also induce a variety of more exotic behaviors and novel quantum phases [4–22], including those that are typically associated with strong repulsive interactions, e.g. anti-ferromagnetism (AF) [10–13] and $d$-wave SC [14–22].

In this letter, we study a simple electron-phonon model, the “Holstein-Lieb” model (illustrated in Fig. 1), for which it is possible to obtain well-controlled results concerning the ground-state phase diagram through the use of an asymptotic strong-coupling expansion. Its schematic quantum phase diagram is summarized in Fig. 2. Certain of the phases are interesting but not surprising – for instance, bond-centered bipolarons can order to form a variety of insulating charge-density-wave states. In the physics of quantum materials, e.g. for Bardeen-Cooper-Schrieffer superconductivity (SC) and typical charge-density-wave ordering [1–3], in the past few years, it has become increasingly clear that electron-phonon interactions can also induce a variety of more exotic behaviors and novel quantum phases [4–22], including those that are typically associated with strong repulsive interactions, e.g. anti-ferromagnetism (AF) [10–13] and $d$-wave SC [14–22].

FIG. 1: An illustration of the Lieb lattices studied in this paper, and the “Holstein-Lieb” model in Eq. 1.
We will perform a controllable strong-coupling analysis assuming $|t|$ to be a small energy scale (the precise meaning of this assumption will be made clear below), and obtain effective theories for the active degrees of freedom in different parameter regimes. Indeed, at the end of the paper we will discuss several aspects of generalizing the model to include direct Hubbard interactions,

$$\hat{H}_{\text{Hub}} \equiv U_c \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + U_f \sum_{\langle ij \rangle} \hat{n}_{\langle ij \rangle\uparrow} \hat{n}_{\langle ij \rangle\downarrow}$$

which can be analyzed in the same controlled manner.

**Methods.** The effects of strong electron-phonon coupling can best be addressed following a unitary transformation

$$\hat{U} \equiv \exp \left[ -\frac{\alpha}{K} \sum_{\langle ij \rangle} \hat{P}_{\langle ij \rangle} \hat{n}_{\langle ij \rangle} \right],$$

that transforms the Hamiltonian into:

$$\hat{U}^\dagger \hat{H} \hat{U} = -t \sum_{\langle ij \rangle} \left[ \hat{D}_{\langle ij \rangle} \hat{f}_{\langle ij \rangle\sigma}(\hat{c}_{\sigma} + \hat{c}_{\sigma}^\dagger) + \text{h.c.} \right] + \sum_{\langle ij \rangle} \left[ \hat{E}_{\langle ij \rangle} \hat{n}_{\langle ij \rangle} - \frac{U_{\text{e-ph}}}{2} \hat{n}_{\langle ij \rangle}^2 \right] + \hat{H}_{\text{ph}}$$

where $\hat{D}_{\langle ij \rangle} \equiv e^{-i\hat{P}_{\langle ij \rangle}\alpha/K}$ is a unitary operator that displaces the phonon coordinate by $\alpha/K$. We will perform a perturbative analysis treating the second line in Eq. 5 as the unperturbed Hamiltonian, $\hat{H}_0$, and the first line as perturbations, $\hat{H}'$. Under most circumstances, $\hat{H}_0$ has an extensive ground-state degeneracy, so we use degenerate perturbation theory to derive an effective model acting in the degenerate subspace. We keep terms in powers of $t$ to the lowest order needed to resolve the degeneracy, and the validity of each model will be analyzed in each scenario. Since $\hat{H}$ can be defined on any lattice in any dimension, so can the resulting effective theories. Here, to be explicit, we will confine ourselves to the square and triangular lattices in two dimensions.

The first step in our analysis is to identify the degenerate ground-state manifold of $\hat{H}_0$, which we call $\mathcal{H}_0$; we will restrict our attention to the range of electron densities per unit cell, $0 < n < 2$ [51]. For convenience, we define $\mathcal{E}_1 \equiv \mathcal{E} - U_{\text{e-ph}}/2$ and $\mathcal{E}_2 \equiv 2\mathcal{E} - 2U_{\text{e-ph}}$ to represent the energies of a singly or doubly occupied bond orbital, and $\mathcal{E}_{12} \equiv \mathcal{E}_1 - \mathcal{E}_2 = 3U_{\text{e-ph}}/2 - \mathcal{E}$ to represent their energy difference. Because $\mathcal{E}_2 - 2\mathcal{E}_1 = -U_{\text{e-ph}}$ is always negative, singly occupied bond orbitals are always disfavored. When $\mathcal{E}_2$ is positive ($U_{\text{e-ph}} < \mathcal{E}$), electrons tend to stay on $c$ (site) orbitals. In this case, $\mathcal{H}_0$ consists of all possible electron occupation configurations on sites. When $\mathcal{E}_2$ is negative ($U_{\text{e-ph}} > \mathcal{E}$), the electrons tend to stay on $f$ (bond) orbitals in pairs; it is convenient to associate a pair of electrons on bond $\langle ij \rangle$ with a dimer occupying this bond. In this language, a basis for $\mathcal{H}_0$ can be labeled by dimer configurations. When $\mathcal{E}_2 \approx 0$, both orbitals are active, the analysis is more complicated as will be discussed later.

For the case where dimers are active degrees of freedom ($\mathcal{E}_2 < 0$), there are two sorts of terms that will be generated by the perturbative analysis: There are diagonal terms (which we will associate with a dimer potential energy) and off-diagonal terms (which we will associate with a dimer kinetic energy). Since the phonon displacements are different for different dimer configurations, all off-diagonal terms must vanish in the limit of non-dynamical phonons, $\omega_0 \rightarrow 0$. Specifically, the amplitude of any process in which a dimer relocates into or out of a bond must be accompanied by a Frank-Condon factor $F$ [26] defined as

$$F \equiv \langle 0 | \hat{D}^2 | 0 \rangle = e^{-X},$$

where $|0\rangle$ is the ground state of the phonon Hamiltonian on the bond, and $X \equiv U_{\text{e-ph}}/\omega_0$ is a dimensionless factor quantifying the degree of retardation, and the displacement operator $\hat{D}$ is squared since the occupancy of the orbital changes by two electrons. This factor becomes arbitrarily small in the limit of strong retardation. On the other hand, the potential terms always only receive $\mathcal{O}(1)$ factors from the virtual phonon fluctuations.
Below and in the Supplemental Materials we derive the effective theories and obtain expressions as functions of the bare energy scales for the coupling constants that arise in low-order perturbation theory in $\tau$. The effective Hamiltonians are given in Eqs. 7, 9 & 11 in terms of the dimensionless parameters $X \equiv U_{\text{e-ph}}/\omega_0$, $Y \equiv \xi_1/\omega_0$, $Z \equiv |E_3|/\omega_0$, and $W \equiv E_{12}/\omega_0$. Each limiting behavior is evaluated in the region of validity of the corresponding effective Hamiltonian.

However, when $X$ is large, such that $F \ll 1$, the $V_1$ term is dominant over $\tau_1$. Those ground states of $V_1$ within $\mathcal{H}_0$ form an emergent low-energy (still extensively degenerate) subspace, $\mathcal{H}_1$, in which, as in the RK quantum dimer models, no more than one dimer can touch a given site. Within $\mathcal{H}_1$, we further perform perturbative analysis on square and triangular lattices and obtain the RK model as the effective model:

$$\hat{H}_{\text{RK}} = V_2 \sum_\sigma \left[ |\sigma\rangle \langle \sigma| + |\bar{\sigma}\rangle \langle \bar{\sigma}| \right] - \tau_2 \sum_\sigma \left[ |\sigma\rangle \langle \bar{\sigma}| + |\bar{\sigma}\rangle \langle \sigma| \right]$$

(8)

where the ket (bra) represents a pair of annihilation (creation) operators on the thickened bonds, and the summation is over all possible four-sided plaquettes. To leading order in $\tau_1/V_1$, it is easy to see that $\tau_2 \approx \frac{4\tau^2}{V_1} \sim \frac{\tau^4}{V_1^4}$.

In terms of the same expansion, one would conclude that $V_2 = \mathcal{O} \left( \tau_1^2 / V_1^2 \right)$ is always small compared to $\tau_2$. However, since we are assuming $F$ is small, we need to consider terms (not shown in Eq. 7) that are higher order in $t$, but which are not suppressed by $F$ (since $V_2$ is a potential term). Since it involves such high-order processes, it is not worth writing out explicitly the resulting expression for $V_2$. What is essential is that it is eighth order in $t$ and positive, i.e. $V_2 \sim \frac{\tau^8}{V_1^4}$. [We have performed exact diagonalization studies on small clusters in the limit $\omega_0 = 0$ as a check of these conclusions.] From the perturbative perspective, there are two types of virtual process that contribute to $V_2$, both of which produce a repulsion: If

| $t_{\text{eff}}$ | $\omega_0 \to \infty$ | $\omega_0 \to 0$ |
|----------------|-----------------|-----------------|
| $\tau_0 = \frac{2t^2}{\xi^2} g_0(Y)$ | $\frac{t^2}{\xi^2} U_{\text{e-ph}}^2/2$ | $\frac{t^2}{\xi^2} U_{\text{e-ph}}^2/2$ |
| $\tau_1 = \frac{2t^4}{\xi^2} \left[ 2g_0^2(W) + \frac{1}{U_{\text{e-ph}}} g_1(W, X) \right]$ | $2t^4 \left( 2U_{\text{e-ph}} - \xi \right) / 4U_{\text{e-ph}}^2$ | $2t^4 \left( 2U_{\text{e-ph}} - \xi \right) / 4U_{\text{e-ph}}^2$ |
| $V_1 = \frac{4t^4}{\xi^2} \left[ \frac{1}{U_{\text{e-ph}}} g_1(W) + \frac{1}{U_{\text{e-ph}}} g_2(W, X) \right]$ | $2t^4 \left( 2U_{\text{e-ph}} - \xi \right) / 4U_{\text{e-ph}}^2$ | $2t^4 \left( 2U_{\text{e-ph}} - \xi \right) / 4U_{\text{e-ph}}^2$ |

TABLE I: Expressions and limiting behaviors of the effective couplings in Eqs. 7, 9 & 11 in terms of the dimensionless parameters $X \equiv U_{\text{e-ph}}/\omega_0$, $Y \equiv \xi_1/\omega_0$, $Z \equiv |E_3|/\omega_0$, and $W \equiv E_{12}/\omega_0$. Each limiting behavior is evaluated in the region of validity of the corresponding effective Hamiltonian.
two dimers occupy two parallel sides of a four-sided plaquette, the lowest order virtual process that non-trivially connects them is a ring exchange of electrons, which results in a fermion minus sign that turns what would have been an energy gain into a cost. If only one dimer occupies a side of a plaquette, there is an allowed virtual process in which a single electron travels around the plaquette - this energy-gaining process is blocked when there are two dimers on the plaquette. [52]

Formally, we can consider $H_{\text{eff}}$ to be the effective Hamiltonian in the asymptotic limit in which $F \sim |t|/\mathcal{E}_2 \sim \delta$ are both small, such that the couplings $\tau_2$ and $V_2$ are both of order $\delta^8$, with relative magnitudes that can be tuned – for instance by varying $\omega_0$ – from a regime in which one is larger to a regime which the other is. All omitted terms are higher order in powers of $\delta$.

The zero-temperature phase diagrams of the quantum dimer models Eq. 8 on the square and triangular lattices have been extensively studied, with results we briefly summarize here. For both cases, $\tau_2 = V_2$ is a special point (RK point [24, 25]) whose ground state is an equal amplitude superposition of all dimer configurations in a given topological sector, i.e. a short-ranged RVB state. On the square lattice, this point is a quantum critical point separating two different VBS states: staggered (for $V_2 > \tau_2 > 0$) and plaquette (for $0 < V_2 < \tau_2$) [33, 34]. On the triangular lattice, the RVB state exhibits $Z_2$ topological order and is stable in a range of $\nu_c J < V_2 < \tau_2$ with $\nu_c \lesssim 0.8$; two different VBS occur for other ranges of parameters: staggered (for $V_2 > \tau_2 > 0$) and $\sqrt{12} \times \sqrt{12}$ (for $0 < V_2 < \nu_c \tau_2$) [35, 36], which we also refer to as “plaquette VBS” in the schematic phase diagram Fig. 2.

The RK point on the square lattice is a strange quantum multi-critical point. A wide class of perturbations can induce incommensurate crystalline phases around the point, through a mechanism known as Cantor deconfinement [37, 38]. We note that these relevant terms generically exist in the higher-order processes that are neglected in this study. It is also worth noting that the inclusion of a small admixture of further (second neighbor) range dimers can stabilize a $Z_2$ spin liquid phase proximate to the RK point, similar to that seen in the triangular lattice [39]. Therefore, slightly away from the asymptotic limit, the RK point is likely to broaden into a regime with multiple additional quantum phases. Interestingly, it is also possible that a power-law critical phase may also be stabilized near a range of parameters around the RK point at finite temperature [40, 41].

The RK point is known to have several types of excitations: spinons, holons, and visions. In the current case, while visions have relatively low creation energy $\sim \tau_2$, the creation of spinons or holons requires large energy cost $\sim \mathcal{E}_2$ or $\sim U_{\text{e-ph}}$ in order to break a dimer. Therefore, when lightly doping holes into the system near the RK point such that $|n-1| = x \ll 1$, we will likely have dimer vacancies as charge carriers leading to condensation with $x \tau_1 \sim x^2 \delta^6$ as the characteristic energy scale (SC transition temperature $T_c$).

$U_{\text{e-ph}} < \mathcal{E}$: weakly interacting model. In this case, the effective model is expressed in terms of site electrons (c-electrons). To fourth order in $t$, the effective Hamiltonian is

$$
\hat{H}_c = -t_{\text{eff}} \sum_{\langle ij \rangle \sigma} \left( \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + \text{h.c.} \right) - 2J_{\text{eff}} \sum_{\langle ij \rangle} \hat{n}_{[i+j]\uparrow} \hat{n}_{[i+j]\downarrow}
$$

(9)

where $\hat{n}_{[i+j]\sigma} \equiv (\hat{c}_{i\sigma}^\dagger + \hat{c}_{j\sigma}^\dagger) (\hat{c}_{i\sigma} + \hat{c}_{j\sigma}) / 2$ is the number of electrons in a bonding orbital between sites $i$ and $j$. Since any virtual movement of electrons necessarily costs $\mathcal{E}_1$ in the intermediate state, the expansion is valid as long as $|t| \ll \mathcal{E}_1$. [Note that, this condition does not require $|t| \ll U_{\text{e-ph}}$.] As can be seen from Table I, $t_{\text{eff}}$ is second order in $t$ and $J_{\text{eff}}$ is fourth order. Thus, we should consider this problem in the “weak coupling” limit, $J_{\text{eff}} \ll t_{\text{eff}}$. This interaction term on each bond can be decomposed into a sum of an AF coupling, a pair hopping term, and a density attraction.

We analyze the weak-coupling instabilities in the context of a Hartree-Fock mean-field analysis that is reasonable in this limit. We find that, on square and triangular lattices, $s$-wave SC is always the dominant instability for $n < 2$. However, when $n \approx 1$ on the square lattice, there is also a $d$-wave pairing state that is meta-stable. To understand the reason for this, one may rewrite the interaction in the form $\sum_{q,k,k'} V_{qkk'} \hat{c}_{q\uparrow k\uparrow}^\dagger \hat{c}_{q\downarrow k'\downarrow}^\dagger \hat{c}_{-q\downarrow -k'\uparrow} \hat{c}_{-q\uparrow -k\downarrow}$. For $q = 0$ and for $k, k'$ near the Fermi surface at $n \approx 1$, we obtain:

$$
V_{k,k'} \approx -2J_{\text{eff}} \left[ 2 \cos k_x c_{k_x} c_{k'_x} + \cos k_y c_{k_y} c_{k'_y} \right] = -2J_{\text{eff}} \sum_{\alpha} f^{(\alpha)}(k) f^{(\alpha)}(k')
$$

(10)

where in the factorized form, the sum runs over an $s$-wave channel ($f^{(s)}(k) = 2$), an extended $s$-wave channel ($f^{(s)}(k) = \cos k_x + \cos k_y$), and a $d$-wave channel ($f^{(d)}(k) = \cos k_x - \cos k_y$). When $n \approx 1$, where the system has a large density of states near the two $X$ points, $(\pi, 0)$ and $(0, \pi)$, of the Brillouin zone of the square lattice, the $d_{x^2-y^2}$-wave channel with form factor $f^{(d)}(k)$ is only moderately subdominant to the dominant $s$-wave channel (which generically is a combination of $s$ and $\bar{s}$). Moreover, the competition between the $s$- and $d$-wave paired states can be tuned by an additional weak on-site Hubbard repulsion, $U_\text{c}$; when $U_\text{c} \gtrsim 2.2 J_{\text{eff}}$, the $d$-wave paired state has the lower variational energy. Exactly at $n = 1$, there is also an AF instability, which is also subdominant to the $s$-wave SC instability, but which is favored over any superconducting state when $U_\text{c} > 2J_{\text{eff}}$.

$U_{\text{e-ph}} \approx \mathcal{E}$: monomer-dimer model. When $\mathcal{E}_2 \approx 0$, both $c$ and $f$ orbitals are active; only states with singly occupied bond orbitals are excluded from the low energy
subspace $\mathcal{H}_0$. More precisely, when $|\mathcal{E}_2| \lesssim \tau_0$ where $\tau_0$ (again given in Table I) is the matrix element for converting a pair of site electrons to a pair of bond electrons, the effective Hamiltonian can be expressed as

$$
\hat{H}_{bc} = \sum_{\langle ij \rangle} \left[ t_{\text{eff}} \left( 2\hat{n}^b_{ij} - 1 \right) 2\hat{n}_{i+j} + (\mathcal{E}_2 - 4t_{\text{eff}}) \hat{n}^b_{ij} \right] + \tau_0 \sum_{\langle ij \rangle} \left[ \hat{b}^\dagger_{ij} \left( \hat{c}^\dagger_{i\uparrow} + \hat{c}^\dagger_{i\downarrow} \right) \left( \hat{c}_{i\downarrow} + \hat{c}_{j\uparrow} \right) + \text{h.c.} \right] \quad (11)
$$

Virtual transitions treated perturbatively cost either $\mathcal{E}$ or $\mathcal{E}_{12}$, so this theory is valid for $|t| \ll \mathcal{E}_1 \approx \mathcal{E}_{12}$.

In the adiabatic limit where $\tau_0 \to 0$, the boson configuration, $\{n^b_{ij}\}$, is non-dynamical and the $c$ electrons are non-interacting. For this problem, we have performed a classical Monte Carlo study to find the ground-state boson configurations on the square and triangular lattices. For both cases, we find that for all $n \leq 2$, the electrons always tend to fill the site orbitals. [For higher densities, we find a phase separation between a dimer-free and a fully packed dimer region, until all the bonds are occupied.] This can be intuitively understood since the first line of Eq. 11 contains a $c$-electron-dimer repulsion. Away from the adiabatic limit, no controllable analysis can be performed, but we nonetheless performed a mean field study treating $b$ and $c$ fields as decoupled. We find an $s$-wave SC phase with $\langle \hat{b}_{ij} \rangle \neq 0$, which presumably connects smoothly to the same phase in $U_{\text{c-ph}} < \mathcal{E}$ case.

It is also interesting to note that there is a hidden symmetry in the original problem when $\mathcal{E}_2 = 0$. We prove this symmetry and its consequences in Supplemental Materials. For arbitrary $t$, when $U_{\text{c-ph}} = \mathcal{E}$ and in the anti-adiabatic limit $\omega_0 \to \infty$, the transformed Hamiltonian in Eq. 5 has a pseudo-spin $SU(2)$ symmetry which is a generalization of that of the Hubbard model on bipartite lattices [42, 43]. This symmetry implies, in the thermodynamic limit, for the ground state of the system at any filling:

$$
\frac{1}{N_{\text{site}}} \left\langle \left| \sum_i \hat{c}_{i\uparrow}^\dagger \hat{c}_{i\downarrow} - \sum_{\langle ij \rangle} \hat{f}_{ij\uparrow}^\dagger \hat{f}_{ij\downarrow} \right|^2 \right\rangle = 0 \quad (12)
$$

If we include $U_c$ and $U_f$, then the condition becomes $\mathcal{E} + U_f/2 = U_{\text{c-ph}} + U_c/2$. There are three possible physical conclusions from this result: 1. The ground state does not have off-diagonal long-range order (ODLRO) [44]. 2. It has uniform ODLRO, but $z\langle \hat{c}_{i\uparrow}^\dagger \hat{c}_{i\downarrow} \rangle = 2\langle \hat{f}_{ij\uparrow}^\dagger \hat{f}_{ij\downarrow} \rangle$ is exact, where $z$ is the number of nearest-neighbor bonds radiating from a site. 3. It has ODLRO, but the pairing order parameters oscillate in space and average to zero, i.e. it is a pair density wave (PDW) state [45]. The possibility that this class of models may host PDW states certainly merits future studies.

**Outlook.** The derivations of the effective theories can be easily generalized to include strong (in comparison to $t$) or even infinite Hubbard repulsion, $U_c$, on the site orbitals. In that case, the model corresponding to $U_{\text{c-ph}} < \mathcal{E}$ at $n \lesssim 1$ will become a $t$-$J$ model (with no double occupancy constraint on site orbitals), albeit with slightly unusual parameters. In particular, the AF coupling $J$ in this model will be enhanced by the effective attraction on the bond orbitals, and an extra nearest neighbor density-density repulsion interaction will be introduced by virtual fluctuations of phonons. On the other hand, the dimer models for $U_{\text{c-ph}} > \mathcal{E}$ are not qualitatively changed by strong repulsive $U_c$. Similarly, as long as $U_f$ is too strong to change $\mathcal{H}_0$, the physics we have discussed will not be qualitatively affected.

In this letter, we have focused on $n \approx 1$ case on triangular and square lattices, but it is clearly worthwhile to extend the analysis to other lattices and to three dimensions. Other interesting avenues for further study include the role of dilute doped holes [46, 47]. Moreover, the present results can provide a useful starting point for numerical studies of the present model at intermediate couplings and arbitrary filling, given that quantum Monte Carlo simulations are free from the notorious fermion sign problem [48]. Most importantly, we hope that the present results suggest new strategies for finding spin liquids and other exotic quantum phases in materials with strong electron-phonon interactions rather than in frustrated quantum antiferromagnets.

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[50] This is reminiscent of recent studies of Ryderberg atom systems [49], in which spin-liquid phases — in the sense of states with topological order and fractionalized excitations — were shown theoretically to arise in systems that are neither dominated by the physics of spins nor related to a putative "Mott insulator."
[51] The full range of possible $n$ runs from 0 to $n = 2 + z$ where $z$ is the number of nearest-neighbor bonds radiating from a site.
[52] As an aside, we note that a $\pi$ external magnetic flux per plaquette will turn this repulsion into attraction.