Centipede ladder at quarter filling

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We study the ground state and excitation spectrum of a quasi one-dimensional nanostructure consisting of a pole and rungs oriented in the opposite directions ("centipede ladder", CL) at quarter filling. The spin and charge excitation spectra are found in the limits of small and large longitudinal hopping $t_\parallel$ compared to the on-rung hopping rate $t_\perp$ and exchange coupling $I_\perp$. At small $t_\parallel$ the system with ferromagnetic on-rung exchange demonstrates instability against dimerization. Coherent propagation of charge transfer excitons is possible in this limit. At large $t_\parallel$ CL behaves like two-orbital Hubbard chain, but the gap opens in the charge excitation spectrum thus reducing the symmetry from $SU(4)$ to $SU(2)$. The spin excitations are always gapless and their dispersion changes from quadratic magnon-like for ferromagnetic on-rung exchange to linear spinon-like for antiferromagnetic on-rung exchange in weak longitudinal hopping limit.

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INTRODUCTION

The spin ladder systems attract much interest in both experimental and theoretical communities during recent years. On the one hand, the ladder systems may be viewed as intermediate configuration between purely one-dimensional systems and periodic arrays of higher dimensions. On the other hand, in many cases the simple theoretical models represent a good prototype for description of fundamental phenomena like metal-insulator transition, formation of various density wave states, low dimensional superconductivity and many other strongly correlated effects. Besides, some of the toy theoretical models are simple enough to be solved exactly on the lattice or in the long wave continuum limit. The important experimental realizations of the ladder systems (see e.g.) renewed an interest to imperfect spin chains and ladders affected by the chemical substitution, pressure or radiation defects. Although the literature on symmetric two-leg ladders at half-filling is quite extensive, not so much is known about the effects of strong asymmetry in the exchange interaction (or hopping) between the legs. Even less is known about the doped two-leg ladders away from the half-filling regime. The most of considered cases are related to so-called incommensurate filling where in spite of strong Coulomb (Hubbard) interaction, the ground state of the ladder is metallic. On the contrary, at the commensurate filling (e.g. quarter filling) the Umklapp processes open a gap in charge excitations spectrum already for weak interaction. These effects can be consistently treated by means of bosonization technique.

In this paper we consider a model characterized by a gap opened due to kinematic constraints even before any interactions are taken into account. This model, which we call a centipede ladder (CL) or single-pole ladder (see Fig.1a,c), can be visualized as the strongly asymmetric limit of the two-leg ladder (Fig.1b) where the interaction along one of the two legs is negligibly small compared to both interaction along the main leg and the rung, e.g. due to spatial orientation of the rungs (see Fig.1a,b).

The family of centipede ladder-type systems schematically presented in Fig.1b was synthesized as a stable organic biradical crystal PNNNO. The analysis of this geometry for the case of half-filling (two electrons on the rung with infinite on-site repulsion) has shown that this model differs from the symmetric two-leg ladder. The half-filled case for the centipede ladder belongs to the specific class of universality manifesting itself in fractional (2/3) scaling of the spin gap. This new scaling was attributed to existence of hidden dynamical symmetries associated with quantum nature of spin-rotators formed at each rung of the ladder and should be contrasted with the integer scaling discovered for the symmetric two-leg ladders. The corresponding model of highly asymmetric two-leg ladder where the interaction between the spins residing at the end of neighboring rungs is zero has been labeled as Spin Rotator Chain model because it may be mapped onto an effective 1D chain with complicated on-site and inter-site exchange interaction revealing "hidden" symmetries of spin rotator.

One may expect that the same geometry of centipede ladder away from the half-filling may also demonstrate a behavior very different from such for symmetric two-leg ladder and/or the partially filled chains. A quarter-filled CL is the next commensurate system where these differences should be looked for. At first sight, the quarter-filled CL, with strictly one electron per rung is equivalent to the quarter-filled two-orbital Hubbard chain (HC) with partially lifted orbital degeneracy. However, additional local symmetries related to the spin and charge degrees of freedom of electrons on a rung influence a structure of the ground state and the spectrum of low-energy excitation and makes the phase diagram of CL
more than that of half-filled HC or quarter-filled
two-orbital HC. The aim of this paper is to reveal these
new features. The phase diagram of CL promises to be
complicated enough, and in this paper we confine our-
selves with several limiting cases.

The manuscript is organized as follows: in Section I
we formulate the model and describe the hierarchy of the
electronic states on the rung. Section II is devoted to
analysis of the limit, where the on-rung ferromagnetic
exchange is dominant. In Section III we consider the
case of weak longitudinal hopping for antiferromagnetic
on-rung exchange. The Section IV is devoted to the case
of strong longitudinal hopping and discussion of various
response functions at quarter filling. Details of calcula-
tions are presented in Appendix. In Conclusions the
summary of results and the perspectives is discussed.

I. MODEL AND HIERARCHY OF RUNG
STATES

The generic model Hamiltonian is \( \mathcal{H} = \mathcal{H}_r + \mathcal{H}_l \), where

\[
\mathcal{H}_r = \epsilon_0 \sum_{si} n_{si} - t_{\perp} \sum_{i} (c_{\alpha i \sigma}^\dagger c_{\beta i \sigma} + \text{H.c.})
\]

\[
\mathcal{H}_l = - \left( t_{\parallel} \sum_{i\sigma} c_{\alpha i \sigma}^\dagger c_{\alpha i+1, \sigma} + \text{H.c.} \right)
\]

is related to the longitudinal electron hopping along the
leg. Here \( s = \alpha, \beta \) enumerates sites belonging to the
leg (\( \alpha \)) and the branch (\( \beta \)) in a given rung \( i \). The
Hamiltonian \( \mathcal{H}_l \) is an extension of the Spin Rotator
Chain Hamiltonian to the case where empty states on the
rungs appear and electron hopping is possible both
within a given rung (\( t_{\perp} \)) and between neighboring rungs
(\( t_{\parallel} \)). We retained here the direct on-rung exchange
interaction \( I_{\perp} \), which can be either ferromagnetic or anti-
ferromagnetic.

It is natural to use a single rung basis for characteriza-
tion of the energy spectrum of SRC. The set \( S \) of 16
rung basis states consists of an empty state \( |0\rangle \) (which
we will call a vacancy), four singly occupied rung states,
four doubly occupied rung states which are split into sin-
glet/triplet configurations, two doubly occupied on-site
states, four triply occupied states and a fully occupied
state with four electrons. In the limit of strong Hubbard
repulsion, \( U \gg t_{\parallel} \), the lowest states on the rung \( i \) belong-
ing to the charge sector \( N_i = 2 \) (doubly occupied rungs)

\[
|2S, i\rangle = \frac{1}{\sqrt{2}}(c_{\alpha i \uparrow}^\dagger c_{\beta i \downarrow}^\dagger - c_{\alpha i \downarrow}^\dagger c_{\beta i \uparrow}^\dagger)|0\rangle
\]

\[
|2T1, i\rangle = c_{\alpha i \uparrow}^\dagger c_{\beta i \downarrow}^\dagger |0\rangle
\]

\[
|2T0, i\rangle = \frac{1}{\sqrt{2}}(c_{\alpha i \uparrow}^\dagger c_{\beta i \downarrow}^\dagger + c_{\alpha i \downarrow}^\dagger c_{\beta i \uparrow}^\dagger)|0\rangle
\]

\[
|2TI, i\rangle = c_{\alpha i +1 \downarrow}^\dagger c_{\beta i +1 \uparrow}^\dagger |0\rangle
\]

Here the quantum numbers of two-electron states are
qualified as a singlet 2\( S \) and triplet 2\( T \lambda \) with \( \lambda = 1, 0, \bar{1} \).

The energies corresponding to the eigenstates \( \mathbf{3} \) are
defined as follows

\[
E_{1,ii} = \epsilon_0, \quad E_{2S, i} = 2\epsilon_0 - j, \quad E_{2T, i} = 2\epsilon_0 - I_{\perp}
\]

where \( |\epsilon_0\rangle \) is a single-electron ionization energy, \( j =
2t_{\parallel}^2/U \) is the indirect exchange of spins at adjacent sites,
which stems from virtual transitions including double oc-
cupancy. Strong Hubbard repulsion effectively excludes
all states beyond the four states \( \mathbf{3} \) in the low-energy
part of the excitation spectrum with \( N_i \geq 2 \) (see below).

In a charge sector \( N_i = 1 \) the electron spectrum demands
further diagonalization. The states on a given rung are characterized by spin and pseudospin. Spin \( s_i \) is
defined as usual, and pseudospin \( \tau_i \) is introduced as

\[
\tau_i^z = \frac{1}{2} \sum_{\sigma}(n_{\alpha i \sigma} - n_{\beta i \sigma}), \quad \tau_i^\perp = \frac{1}{2} \sum_{\sigma} [c_{\beta i \sigma}^\dagger c_{\alpha i \sigma} + \text{H.c.}]
\]

The kinetic energy term in \( \mathcal{H}_l \) corresponding to the hopping
along the rung may be cast in the form

\[
\mathcal{K}_{\perp} = -2t_{\perp} \sum_i \tau_i^z
\]

while, assuming two different electro-chemical potentials
\( \mu_{\alpha} \) and \( \mu_{\beta} \) (or, equivalently, two different surround-
ings for \( \alpha, \beta \) sites), we get one more term

\[
\mathcal{K}_\mu = - \sum_i \tau_i^z (\mu_{\alpha} - \mu_{\beta} - \frac{1}{2} \sum_i N_i (\mu_{\alpha} + \mu_{\beta})
\]
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The lowest excited state is

\[ |\pm, \sigma, i\rangle = \frac{1}{\sqrt{2}} (|1, \alpha i\sigma\rangle \pm |1, \beta i\sigma\rangle) \]  

(8)

At finite \( t_\perp \) this basis still diagonalizes the rung Hamiltonian and the corresponding single-electron energy levels are \( \epsilon_{\pm} = \epsilon_0 \mp t_\perp \). One should stress that the doubly occupied states cannot be viewed as two bonding and/or antibonding orbitals on a rung, due to strong Hubbard repulsion, and the charge sector \( N_i = 2 \) is represented by the states (3). This is a specific property of CL in comparison with the two-orbital HC.

Another remarkable property of CL which, in fact, allows double occupation of a rung without paying large energy \( U \), is its instability against chain dimerization under certain conditions. To describe this instability, one should introduce the elementary cell for a CL with doubled lattice period. Such cell is occupied by two electrons, and its energy spectrum at zero \( t_\parallel \) is easily found. The low-energy subset is formed by the dimerized states and the states with singly occupied rungs, respectively:

\[
\begin{align*}
\{ N_i = 2, N_{i\pm 1} = 0 \} &= \{|2T\lambda, i; 0, i + 1\}, |2S, i; 0, i + 1\} \\
\{ N_i = 1, N_{i\pm 1} = 1 \} &= \{|\pm, \sigma, i; \pm, \sigma', i + 1\}\end{align*}
\]

(9)

The corresponding two-electron energy levels are

\[
\begin{align*}
E_T(2, 0) &= 2\epsilon_0 - I_\perp \\
E_S(2, 0) &= 2\epsilon_0 - j \\
E_{++}(1, 1) &= 2(\epsilon_0 - t_\perp) \\
E_{+-}(1, 1) &= 2\epsilon_0 \\
E_{--}(1, 1) &= 2(\epsilon_0 + t_\perp)
\end{align*}
\]

(10)

One concludes from (10) that the lowest energy levels which predetermine the spectrum of quarter-filled CL are \( E_T(2, 0), E_S(2, 0), E_{++}(1, 1), E_{--}(1, 1) \). If \( I_\perp < 2t_\perp \), the lowest two-electron states of this elementary cell belong to the subset \( \mathcal{S}_H = \{+, \sigma, i; +, \sigma', i + 1\} \) with one bonding orbital per rung. In the opposite case \( I_\perp > 2t_\perp \) the lowest state correspond to (2,0) occupation of adjacent rungs. We denote the corresponding subset as \( \mathcal{S}_d = \{2T, 2S, i; 0, i + 1\} \). At finite longitudinal hopping CL forms homogeneous quarter-filled chain in the former case and dimerized chain with alternating (2,0) occupation in the latter case.

In case of negative (antiferromagnetic) exchange \( I_\perp \) the ground state is homogeneous quarter-filled chain belonging to the subset \( \mathcal{S}_H = \{+, \sigma, i; +, \sigma', i + 1\} \) and the lowest excited state is \( E_S \) (see Fig. 2b).

In two following sections we discuss charge and spin excitation spectra of CL in the limit of weak longitudinal hopping. Looking at Fig. 2b, c and Eqs. (10), we see that in case (a) one may find a regime, where only the levels \( E_{++}, E_T \) are involved in formation of the low-energy excitations, whereas in case (b) at least three states \( E_{++}, E_S, E_{--} \) are involved.

FIG. 2: (Color online) Two electron level structure for two neighboring rungs. a) Ferromagnetic on-rung exchange. The ground state is dimerized. b) Antiferromagnetic on-rung exchange. The ground state is array of bonding orbitals. c) Occupation scheme for two different ground states: filled, empty and half-filled circles correspond to occupied (triplet), empty (vacancy) and bonding electron states respectively. Transitions within a given charge spin or charge sector and transitions between different sectors are marked by vertical and slanting dashed arrows, respectively. The energies of this transitions are also shown.

II. WEAK LONGITUDINAL HOPPING LIMIT, FERROMAGNETIC ON-RUNG EXCHANGE

In this section we analyze the quarter-filled CL with \( I_\perp > 0 \) in a situation, where the longitudinal hopping is small in comparison with the on-rung coupling constants, and the inequalities

\[ I_\perp > 2t_\perp, \quad t_\parallel \ll I_\perp - 2t_\perp \]

(11)

are valid. In the absence of longitudinal hopping each second rung is empty and all doubly occupied rungs are
in spin one triplet states (see Fig. 2a). One has to study longitudinal spin and charge excitations at finite \( t_\parallel \) above the dimerized ground state with alternating rungs \( \ldots |2T\lambda, i⟩, |0, i+1⟩ \ldots \). The charge excitations arise due to transitions \( S_d \leftrightarrow S_h \). These excitations (charge-transfer excitons) are obviously gapped, so that the low-energy excitations belong to the spin sector.

In zeroth order approximation the spectrum of rung dimers is determined by the eigenstates of the Hamiltonian (1) in a charge sector \( N_i = 2 \) for FM on-rung exchange. These dimers are in the state of spin rotator \( 2 \) characterized by the singlet-triplet manifold \( \{ |2T\lambda, i⟩, |2S, i⟩ \} \), with the energy gap between two spin states given by

\[
\Delta E_{ST} = I_\perp - j. \tag{12}
\]

Longitudinal hopping (2) generates indirect exchange produced by the electron cotunneling via empty odd rungs. To derive the corresponding spin Hamiltonian, we have to exclude the charge sector \( S_h \) from the effective phase space. It is seen from Fig. 2a, that the singlet states are effectively quenched at the energy scale \( I_\perp - 2t_\perp \), which characterizes the charge transfer gap, so that one may confine oneself with the subspace \( \{ S'_d, S_h \} \), with singlet states excluded from the subset \( S'_d = \{ 2T, i; 0, i+1 \} \) of ”polar” states.

The remarkable property of the state \( S'_d \) is that the macroscopic configurational degeneracy of the Hubbard model with partial filling is removed due to dimerization. The only remaining even-odd degeneracy of the dimerized ground state of zero-order Hamiltonian may be also removed in case of CL with odd number of rungs \( M = 2N + 1 \), where, say, all even rungs 0, 2, \ldots \( 2N \) are doubly occupied and all odd rungs 1, 3, \ldots \( 2N - 1 \) are empty. Then only the trivial spin degeneracy remains, and one may strictly define the effective exchange Hamiltonian in the 4-th order of Brillouin-Wigner expansion

\[
\mathcal{H}_{\text{eff}} = \mathcal{H}_r + \langle \Psi_0 | \mathcal{H}^{(4)}_l | \Psi_0 \rangle, \tag{13}
\]

where the brackets \( \langle \Psi_0 | \ldots | \Psi_0 \rangle \) stand for the dimerized polar ground state with alternating triplet spin states and vacancy states

\[
\mathcal{H}^{(4)}_l = \frac{\mathcal{H}_l |\phi^{(1)}⟩⟨\phi^{(1)}| + |\phi^{(2)}⟩⟨\phi^{(2)}| + |\phi^{(3)}⟩⟨\phi^{(3)}|}{(−E^{(1)}_0)(−E^{(2)}_0)(−E^{(3)}_0)} \tag{14}
\]

where

\[
P^{(\alpha)} = |\phi^{(\alpha)}⟩⟨\phi^{(\alpha)}|, \quad \alpha = 1, 2, 3
\]

include the states which are generated from the ground state \( |\Psi_0⟩ \) after 1-st, 2-nd and 3-rd action of the operator \( \mathcal{H}_l \), respectively and \( E^{(\alpha)}_0 \) are the corresponding excitation energies. The 4-th order cotunneling process resulting in effective kinematic exchange is illustrated by Fig. 3. It is evident that only the clusters \( (i, i+1, i\pm 2) \) are involved in formation of the effective exchange Hamiltonian.

\[
H_{\text{eff}} = H_r - J∥ \sum_i S_i \cdot S_{i+2} \tag{16}
\]

where \( i \) stands for any even site. We consider here the simplest situation, where the singlet intermediate states can be discarded in the main order, which happens at \( t∥/(I_\perp - j) \ll t∥/(I_\perp - 2t_\perp) \). This simplification does not change our qualitative conclusions.

\[
|\phi^{(1)}⟩ = | + σ_1, i⟩ + σ_2, i + 1⟩ + 2Tλ, i + 2⟩
\]

\[
|\phi^{(2)}⟩ = | + σ_1, i⟩ + 2Tλ_1, i + 1⟩ + + σ_3, i + 2⟩
\]

\[
|\phi^{(3)}⟩ = | + σ_1, i⟩ + σ_3, i + 1⟩ + 2Tλ', i + 2⟩ \tag{15}
\]

It is seen from Eq. (15) and Fig. 3 that the intermediate states involve electron cotunneling exchange between two nearest even sites with formation of a virtual doubly occupied triplet state on the odd site between them. This mechanism is known as superexchange in the theory of magnetic dielectrics. Appearance of intermediate triplet states \( |2Tλ⟩ \) in the indirect exchange interaction is the peculiarity of CL: unlike the two-orbital HC model, double occupation of intermediate state does not cost the Hubbard repulsion energy \( U \), because the two electrons on a rung may occupy different sites. As a result we come to the following effective spin Hamiltonian

\[
\mathcal{H}_{\text{eff}} = \mathcal{H}_r + \frac{t^4_∥}{16Δ_T^2} \tag{17}
\]

where \( Δ_T = I_\perp - 2t_\perp \) is the charge transfer energy gap (see Fig. 2a). The details of calculation are provided in Appendix. Deriving (17) we assumed that the singlet state does not contribute to superexchange. This assumption holds provided \( Δ_T \ll Δ_{ST} \).

Thus we have demonstrated that the problem of quarter-filled CL is mapped onto the familiar problem of spin-one Heisenberg chain with ferromagnetic exchange in the limit of weak longitudinal hopping under conditions (11). Only the even sites are involved in formation of the gapless spin excitations, which, apparently may be described in terms of the spin-wave theory.
As to the charge transfer excitons, \(| + \sigma, i; + \sigma', i + 1 \rangle\), shown in the right column of Fig. 2c, these states, like other excitons can exist both in triplet and singlet spin configurations. These excitations may propagate coherently by means of cotunneling reminding that described by Eqs. (14), (15). To provide translation of such exciton \(\epsilon_{\pm}(k)\) are shown. The chemical potential is chosen as a reference energy level.

Thus, we have shown in this section that the spin excitations in the CL with ferromagnetic on-rung exchange are gapless magnon-like modes, whereas the excitations in a charge-sector are gapwise excitons describing coherent propagation of two bonding states along the chain.

III. WEAK LONGITUDINAL HOPPING LIMIT, ANTIFERROMAGNETIC ON-RUNG EXCHANGE

In case of negative on-rung exchange hopping along the leg and small \(t_{\perp}\), so that the inequality

\[
t_{\parallel} \ll 2t_{\perp} - j
\]

is satisfied, the energy levels are ordered in accordance with Fig. 2a, and the ground state wave function of quarter-filled CL is given by the product of bonding orbitals

\[
\langle \Psi_{0} \rangle = | \ldots + \sigma_{i}, i; + \sigma_{i+1}, i+1; + \sigma_{i+2}, i+2; \ldots \rangle.
\]

In this sector of phase diagram, the quarter-filled two-orbital HL is close in a sense to the quarter-filled two-orbital HL with lifted orbital degeneracy: each rung is occupied by one electron with bonding orbital, and the empty antibonding level is separated from this band by the orbital gap \(\sim 2t_{\perp}\) [see Eqs. (9), (10) and Fig. 4].

The important distinction from two-orbital HL roots in the structure of the doubly occupied states. Since two electrons on a rung occupy different orbitals in accordance with (11), the energy cost of this state is not the Hubbard repulsion energy \(U\) but the difference \(\Delta_{S} = E_{S} - E_{\perp} = 2t_{\perp} - j\), which is even smaller than the orbital gap. Nevertheless, magnetic properties of these two models are similar in the limiting case (15): the longitudinal hopping generates effective AFM longitudinal exchange \(\sim t_{\parallel}^{2}/\Delta_{S}\), and the gapless spin liquid state of the resonance valence bond (RVB) type is realized.

The charge excitations exhibit more interesting character, as we discuss below. The lowest excited state is a vacancy-singlet pair occupying neighboring rungs

\[
|S, i; 0, i + 1 \rangle = | \ldots + \sigma, i - 1; - \sigma, i; 0, i + 1; + \sigma, i + 2 \ldots \rangle
\]

with the energy \(\Delta_{S}\).

Clearly, the state \(|S, i; 0, i + 1 \rangle\) is an analog of the charge transfer exciton briefly discussed in the end of section II. However, in this case the exciton propagates via intermediate bonding-antibonding levels \(\epsilon_{\pm}\) due to the strong inequality \(t_{\perp} \gg j\). Figure 5 illustrates the mechanism of exciton propagation. Here the rung occupied by the antibonding electron is shown by the pair of half-filled circles with filled upper half. Then the elementary hopping from doubly occupied to empty rung may be presented as

\[
|S, i; 0, i + 1 \rangle \rightarrow | \pm, i; \mp, i + 1 \rangle,
\]

and it costs the energy \(j\), according to (10). Note that a crucial detail allowing one to discuss exciton propagation as the charge excitation is the fact, that the spin sequence of the bonding orbitals in the initial and final state in Fig. 5 is unchanged and the spinon dispersion is unaffected by the coherent motion of the singlet exciton in the long
wave limit. This is a manifestation of the spin charge separation in quarter-filled CL model.

In the limit of extremely weak longitudinal hopping \( t_{\parallel} \ll j \), the singlet-vacancy pair may propagate coherently. This pair is doubly degenerate due to its permutation symmetry, and this degeneracy manifests itself in existence of right- and left-moving charge-transfer excitons. The 4-th order hopping process resulting in translation of the right-mover between two adjacent double cells is shown in Fig. 4. Superposition of right- and left-moving excitons results in the dispersion law

\[
\omega_q \sim \Delta_S - J'_{\parallel} \cos 2qa
\]

with \( J'_{\parallel} \sim t_{\parallel}^4/j^3 \). We showed before that the spin sequence remains unchanged (Cf. Ref[1]) upon the motion of exciton, and the gain in the kinetic energy is \( \sim J'_{\parallel} \). The resulting spin configuration in Fig. 5 corresponds to two spins detached from the main spin sequence on the right and attached to one on the left. For long-wave modes of exciton motion, \( q \rightarrow 0 \) in (22), the change in the magnetic associated with this disruption is negligible compared to the gain \( \sim J'_{\parallel} \), and charge excitons move coherently.

Now we turn to the case \( t_{\parallel} > j \). In this situation the coherent "vacancy-singlet" excitons are unstable against dissociation into independently propagating singlet and vacancy "defects" due to the overlap of the bands originating from the levels \( E_S \) and \( E_{\perp} \). Apparently, only incoherent propagation of such defects is possible. An example of 4-th order process resulting in decay of singlet-vacancy pair is presented in Fig. 6. Such incoherent processes will be accompanied by spin-flips, antibonding-bonding de-excitation and eventual relaxation towards the ground state.

However, coherent motion of defects is possible at small deviation from one-quarter filling. Excess holes (vacancies) behave similarly to the holes in the nearly half-filled HC in the limit [13], where the longitudinal hopping does not result in excitation of the upper orbital subband \( \epsilon_\perp \). In this case one deals with a fermion gas, “fully polarized” with respect to the pseudospin \( \tau \). Excess electrons fill the side sites on the rungs without paying the Hubbard energy \( U \), and it is easy to check that the hopping along the leg is not accompanied by creation of a spin string because the doubly occupied state is a spin singlet. As a result, the coherent propagation of these defects is also possible under the conditions [13], when the antibonding orbitals are not excited. Due to specific geometry of CL, there is no particle-hole symmetry near the 1/4 filling, and the hopping rates for particle and hole propagation are different. Direct calculation shows that the hopping rate for coherent motion of a vacancy reduces due to the kinematic constraint from \( t_{\parallel} \) to \( \tilde{t}_{\parallel} = t_{\parallel}/2 \), whereas the similar reduction for the singlets gives \( \tilde{t}_s = t_{\parallel}/4 \). Thus, basically the coherent motion of the vacancy and singlet can be considered in terms of tight-binding bands \( \epsilon_{1,2}(k) = -2\tilde{t}_{1,2} \cos k \). Evidently, singlet (vacancy) can-not occupy the same rung twice, hence the corresponding excitations are fermion-like. We can think of them as of spinless fermions characterized by two colors \( f_{i,v} \) and \( f_{i,s} \):}

\[
|0,i\rangle\langle +,i| \rightarrow f_{i,v}^\dagger, \quad |S,i\rangle\langle +,i| \rightarrow f_{i,s}^\dagger \quad (23)
\]

Even at exact quarter filling but at finite temperatures, the vacancies and singlets may appear at small concentrations due to thermal activation. These excitations are described by the effective two color Fermi liquid model

\[
H_{\text{eff}} = \sum_{k,\alpha = v,s} \epsilon_{\alpha}(k) f_{k,\alpha}^\dagger f_{k,\alpha} + K \sum_{j} f_{j,v}^\dagger f_{j,v} f_{j,s}^\dagger f_{j,s} \quad (24)
\]

where the effective constant \( K \) describes effective short-range repulsion between two fermions, which prevents them from occupying the same site. This constant may be estimated as \( K \sim t_{\parallel}^2/\Delta_S \), which is small comparing to the band-widths \( \sim t_{\parallel} \). Having in mind the asymmetric hopping Hubbard model[20], it is tempting to analyze it using the well-established machinery of bosonization etc. However, the sizeable concentrations of vacancies and singlets are expected at temperatures comparable to the main parameter, \( t_{\parallel} \). This argument shows that that fermion gas in \( H_{\text{eff}} \) is not degenerate, and the linearization of the spectrum around Fermi points is unjustified.

As to the excitations in the spin sector, one may say that the problem of spin excitations in presence of a singlet or a vacancy may be mapped on the problem of RVB spin liquid with spin-one defect or dangling bond, respectively (see Ref. [16 for general approach]).

To summarize, we have found in this section, that the spin excitations are gapless Fermi-like spinons both at exact 1/4 filling and at small deviation from this point. As to the coherent charge excitations, these are gapped.
excitons at extremely small $t_j \ll j$ and gapless fermions at finite doping and/or finite temperature.

IV. STRONG LONGITUDINAL HOPPING

The CL model possesses unusual properties also in the limit of strong longitudinal hopping, when it dominates over the transversal degrees of freedom:

$$t_{\|} \gg t_{\perp} \gg I_{\perp},$$

(25)

In this case the on-rung excitations shown in Fig. 1b,a, respectively. Fourier transforming along the leg, we write the kinetic part of the Hamiltonian as

$$H_{\text{kin}} = -\Psi_k^\dagger \left[ 2t_{\|} \cos k + \mu , t_{\perp} , 2Pt_{\perp} \cos k - \mu \right] \Psi_k$$

(26)

$$\Psi_k = (c_{\alpha,k,\sigma}^\dagger , c_{\beta,k,\sigma}^\dagger )$$

(27)

where $P = 1$ for the 2L model and $P = 0$ for CL model. At this point we also allow for the difference in the electrochemical potentials, $2\mu = \mu_\alpha - \mu_\beta$, between the sites $\alpha$ and $\beta$ [see Eq. 4]. The dispersion in these two extreme cases is given by

$$\epsilon_{\pm}(k) = -2t_{\|} \cos k \mp \sqrt{\mu^2 + t_{\perp}^2}, \quad \text{2L}$$

(28)

$$= -t_{\|} \cos k \mp \sqrt{t_{\perp}^2 + (\mu + t_{\perp}) \cos k}^2, \quad \text{CL}$$

(29)

The square root term in Eq. 28 has a meaning of effective Zeeman splitting. In contrast to 2L situation where the bands $\epsilon_+(k)$ and $\epsilon_-(k)$ bands overlap we see that in CL case two branches of dispersion are always separated by the gap (Fig. 1). This indirect gap is given by

$$\Delta_{\text{ind}} = 2(\sqrt{t_{\perp}^2 + t_{\|}^2} - t_{\|}) \simeq \frac{t_{\perp}^2}{t_{\|}},$$

(30)

for the case of equal electrochemical potentials ($\mu = 0$) or

$$\Delta_{\text{ind}} \simeq \frac{t_{\perp}^2 t_{\|}}{(t_{\perp}^2 - \mu^2)}$$

(31)

for $\mu_\alpha \neq \mu_\beta$. The direct gap is realized at $k = \pi/2$ for $\mu = 0$ and at $k = \arccos(-\mu/t_{\perp})$ otherwise and is equal to $\Delta = 2t_{\perp}$ in both cases. For exact 1/4 filling, the overall chemical potential $\mu_0 = \mu_\alpha + \mu_\beta$ lies within a gap separating two bands. The existence of two (direct and indirect) gaps makes the CL case principally different from 2L model.

One could expect that in the limit $t_{\|} \gg t_{\perp}$ the properties of the system will be close to those of two-orbital Hubbard chain\cite{11,21} because the orbital degeneracy is nearly removed by strong longitudinal hopping. This is not the case, however. The gap in the spectrum prevents formation of $SU(4)$ manifold formed by spin and orbital degrees of freedom. However the interband transitions as well as the edge singularities in the DoS influence the thermodynamic and optical properties of CL

In the remainder of this section we concentrate on the case $\mu = 0$ and consider the manifestation of singularities in the excitation spectrum in different response functions of CL. The density of states $\rho(E) = (2\pi)^{-1} \int dk \delta(E - \epsilon(k))$ is divergent at $E = \pm t_{\perp}$ and at $E = \pm (t_{\|} + \sqrt{t_{\perp}^2 + t_{\perp}^2})$. It behaves as

$$\rho(E) \propto \Delta_{\text{ind}}^{-1/2} \sum_{\pm} \frac{\vartheta(-\Delta_{\text{ind}} \pm E)}{\sqrt{\pm E - \Delta_{\text{ind}}}}$$

(32)

at small energies $|E| \sim \Delta_{\text{ind}}$; here $\vartheta(x) = 1$ at $x > 0$. Such character of the density of states (see inset in Fig. 7) should lead to observable anomalies in the thermodynamic quantities. The specific heat $C$, compressibility $\chi$ and dc conductivity $\sigma_{dc}$ are sensitive to indirect gap. Setting all fundamental constants to unity we write for
thermodynamic quantities per unit cell
\[ C = \int \frac{E^2 dE}{4T^2 \cosh^2 (E/2T)} \rho(E) \]
\[ \sim \left( \frac{T}{\Delta_{\text{ind}}} \right)^{1/2}, \ T \gg \Delta_{\text{ind}} \]
\[ \sim \left( \frac{\Delta_{\text{ind}}}{T} \right)^{3/2} e^{-\Delta_{\text{ind}}/T}, \ T \ll \Delta_{\text{ind}} \] (33)

for the specific heat and
\[ \chi = \int \frac{dE}{4T \cosh^2 (E/2T)} \rho(E) \]
\[ \sim (T\Delta_{\text{ind}})^{-1/2}, \ T \gg \Delta_{\text{ind}} \]
\[ \sim (\Delta_{\text{ind}} T)^{-1/2} e^{-\Delta_{\text{ind}}/T}, \ T \ll \Delta_{\text{ind}} \] (34)

for the compressibility along the ladder. Assuming the elastic quasiparticle lifetime \( \tau_{el} \), the dc conductivity is given by
\[ \sigma_{dc} = \tau_{el} \int \frac{dk}{4T \cosh^2 (\varepsilon_{\pm}(k)/2T)} \left( \frac{d\varepsilon_{\pm}(k)}{dk} \right)^2 \]
\[ \sim \tau_{el} \sqrt{T \Delta_{\text{ind}}}, \ T \gg \Delta_{\text{ind}} \]
\[ \sim \tau_{el} \sqrt{T \Delta_{\text{ind}} e^{-\Delta_{\text{ind}}/T}}, \ T \ll \Delta_{\text{ind}} \] (35)

In contrast to behavior of thermodynamic quantities sensitive to indirect gap, the optical conductivity response is sensitive to direct gap. It is non-zero at \( |\omega| > \Delta \) and is given by22:
\[ \sigma_{opt}(\omega) = \frac{1}{\omega} \int dk [n(\varepsilon_-(k)) - n(\varepsilon_+(k))] \]
\[ \times t_1^2 t_1^2 \sin^2 k \]
\[ \times t_1^2 + t_1^2 \cos^2 k \delta(\omega + \varepsilon_-(k) - \varepsilon_+(k)) \]
\[ \sim \frac{t_1^2 \Delta^2}{\omega^2 / \omega^2 - \Delta^2}, \ T \ll \Delta \]
\[ \sim \frac{t_1^2 \Delta^2}{|\omega| T \sqrt{\omega^2 - \Delta^2}}, \ T \gtrsim \Delta \] (36)

We thus see a continuous absorption band in \( \sigma_{opt}(\omega) \) in contrast to the case of 2L ladder, where the optical conductivity consists of a single line, \( \sigma_{opt}(\omega) \propto \delta(\omega \pm \Delta) \).

Interband optical transitions may result in creation of excitons with a center-of-mass momentum \( Q = \pi \). Unlike the case of weak longitudinal hopping (section II), these excitons arise in the conventional way due to attractive Coulomb interaction between electrons and holes.

The bound electron-hole states formed on the almost flat parts of the spectrum over the indirect gap are characterized by heavy effective mass. We therefore expect that these excitons are nearly localized and one may neglect the processes of their coherent motion.

V. CONCLUSIONS AND PERSPECTIVES

In this paper we have demonstrated several fragments of the rich phase diagram for centipede ladder. It was shown in our previous studies10,11 that the half-filled CL is an example of spin 1 chain with soft triplet-singlet excitation, which does not belong to the Haldane gap universality class. Now we have found that the 1/4 filled CL also demonstrate unconventional properties both in spin and charge sectors.

We considered here only three limiting cases of the quarter filled CL where the character of charge and spin excitations may be revealed by means of relatively simple arguments. First, we have found the regime where CL behaves as a spin 1 chain with ferromagnetic effective longitudinal exchange and magnon-like excitation spectrum (Section II). It is shown that the system is unstable against dimerization at strong enough on-rung ferromagnetic exchange. The only coherent mode in the charge sector is the gapped triplet charge-transfer exciton.

One should stress that the dimerisation effect offered in this section has no analogs in current literature. Usually the dimerization mechanism in spin chains is either explicitly built in the Heisenberg Hamiltonian or arises as a result of competition between the nearest and next nearest exchange coupling23. As to the two-leg ladders, the dimerized configurations were considered in a context of RVB states (rung-dimer states vs leg-dimer states)24. In our case the lattice period is doubled as a result of competition between the on-run hopping and exchange coupling. Experimentally, rung dimerization as a result of charge redistribution between legs and rungs was detected optically26 in two-leg compounds AV6O15 (A=Na,Sr). However, in vanadium bronzes this effect is driven mainly by the valence instability of V ions.

Second, we have described the situation, where the vacancies (empty rungs) and spin singlets (doubly occupied rungs) may propagate either as coherent excitons or as independent spinless fermionic quasiparticles with different Fermi velocities (Section III). The dispersion law is gapped in the former case and gapless in the second case, where the charge sector may be described as a two-component Fermi liquid with weak short-range interaction. The spin subsystem behaves as the RVB-type spin liquid, where the spinons propagate in presence of spin 1 and dangling bond defects.

Third, we have considered the limit of strong longitudinal hopping, where the CL behaves as a specific version of two-orbital Hubbard chain. The peculiar features of this limit is the appearance of a gap in the half-occupied lower Hubbard band. This gap has purely hybridization nature, but the van-Hove singularities in the density of states strongly influence the thermodynamic and optical properties of CL.

Our analysis of these three cases by no means exhausts the variety of unusual properties of CL at partial filling. In particular, we didn’t include in our Hamiltonian direct longitudinal exchange which can enhance...
the indirect kinetic exchange or compete with it. We didn’t consider the phase transitions between different phases, which may have their own peculiarities as compared to currently studied quantum phase transitions in low-dimensional systems. We also didn’t discuss possible lattice distortions, which may accompany dimerization. All these and many other open questions are left for future investigation.

The centipede ladder shown in Fig. 1 is the simplest example of the family of quasi 1D systems intermediate between chains and two-leg ladders. The experimental realization of such system (organic biradical crystals PNNNOx) is already known, but one may easily imagine molecular chains decorated not with rungs but with radicals forming closed loops, zigzags etc. We believe that the study of excitation spectra, magnetic and transport properties of this family will bring new unexpected results.

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APPENDIX: CALCULATION OF THE SUPEREXCHANGE INTERACTION

In order to calculate $J_∥$ we note that two spins $S = 1$ at even sites in Eq. (16) and in Fig. 3 can be combined into total spin $S_{tot} = 0, 1, 2$. The scalar product $S_i \cdot S_{i+2} = \frac{1}{2}(S_i^2 + S_{i+2}^2 - S_i^0 - S_{i+2}^0)$ in (16) attains values $-2$, $-1$, $1$ for these values $S_{tot}$, respectively. The value of $3J_∥$ can hence be naively calculated as a difference in fourth-order corrections $[\Phi]$ to the states $S_{tot} = 0$ and $S_{tot} = 2$.

Let us use a shorthand notation $|m, m\prime\rangle$ for a quantum state $|Tm, i; Tm', i + 2\rangle$, and $\Phi_{L,m}$ for the state with total spin $S_{tot} = L$ and its $z$–component $m$. The weight of $\Phi_{L,m'}$ in $|m, m\prime\rangle$ is the Clebsch-Gordan coefficient $C_{1m1m'}^L$. Then the states we are interested in, $S_{tot} = 0$ and $S_{tot} = 2$, are given by $\Phi_{0,0} = (|−1, 1\rangle + |1, −1\rangle − |0, 0\rangle) / \sqrt{3}$ and $Φ_{2,2}|1, 1\rangle$.

Further, we denote the combinations $|+\sigma_1, i; +\sigma_3, i + 2\rangle$ in $|φ^{(2)}\rangle$, Eq. (15) as ”non-local triplet” states, e.g. $|+\uparrow, i; +\uparrow, i + 2\rangle = |T\bar{1}, i + 1\rangle$ etc. The intermediate state in Fig. 3, obtained after two longitudinal hops in terms of its spin components, is rather obviously classified in the same terms of total spin, composed of two triplets, $|Tm, i + 1\rangle$ and $|T\bar{m}, i + 1\rangle$. Similarly denoting $|T\bar{m}, i + 1; Tm', i + 1\rangle = |m, m\prime\rangle$ we have $\Phi_{0,0} = (|−1, 1\rangle + |1, −1\rangle − |0, 0\rangle) / \sqrt{3}$ etc.

It then can be shown that (projecting at each step at the states with lowest energy, bonding and triplet, as discussed above)

$$H_7|−1, 1\rangle = \frac{t_∥^2}{2\sqrt{2}}|\downarrow, i; T0, i + 1; +\uparrow, i + 2\rangle$$

$$H_7(|−1, 1\rangle + |1, −1\rangle) = \frac{t_∥^2}{2}|0, 0\rangle = \frac{t_∥^2}{2}\sqrt{\frac{2}{3}}\tilde{\Phi}_{2,0} - \sqrt{\frac{2}{3}}\tilde{\Phi}_{0,0}$$

$$H_7|0, 0\rangle = \frac{t_∥^2}{4}(|−1, 1\rangle + |1, −1\rangle + |0, 0\rangle) = \frac{t_∥^2}{4}[2\sqrt{\frac{2}{3}}\tilde{\Phi}_{2,0} + \sqrt{\frac{1}{3}}\tilde{\Phi}_{0,0}]$$

(A.1)

which leads to $H_7^2\Phi_{0,0} = −\frac{t_∥^2}{2}\tilde{\Phi}_{0,0}$. Similarly, one obtains $H_7^2\Phi_{2,2} = \frac{t_∥^2}{2}\tilde{\Phi}_{2,2}$. Because the projected action of $H_7^2$ is self-conjugate, we obtain $H_7^4\Phi_{0,0} = t_∥^4\Phi_{0,0}$ and $H_7^4\Phi_{2,2} = t_∥^4\Phi_{2,2}$.

We would also like to exclude more complex structure of the effective Hamiltonian, namely

$$H_{eff} = −J_∥\sum_i (S_i \cdot S_{i+2} + c_1(S_i \cdot S_{i+2})^2)$$

(A.2)

which is not prohibited by symmetry.

To this end, we calculate also the correction to states $\Phi_{1,m}$ of total spin $L = 1$. Within the same logic, we have $H_7^2(|1, −1\rangle − |−1, 1\rangle) = H_7^2\sqrt{2}\tilde{Φ}_{1,0} = \frac{t_∥^2}{2}|\tilde{S}, i + 1; T0, i + 1\rangle$ where $\tilde{S}$ non-local singlet. Therefore $H_7^4\Phi_{1,0} = \frac{1}{8}t_∥^4\Phi_{1,0}$ and for our values $L = 0, 1, 2$ we have the corrections $−1/16, −1/8, −1/4$ in units of $t_∥^4/Δ_∥^4$, respectively. It means that $J_∥ = t_∥^4/(16Δ_∥^4)$ and $c_1 = 0$. Thus the anisotropy term $(S_i \cdot S_{i+2})^2$ does not appear in the forth order of longitudinal hopping. The reason of it is very simple: in order to get the four-spin $S = 1$ interaction within the scheme shown on Fig. 3 where four spins $s = 1/2$ participate, one needs at least 8 processes of longitudinal hopping.
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