On charge and spin ordering in a one-dimensional model with frustrating interactions.

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Abstract. – We study a one-dimensional extended Hubbard model with longer-range Coulomb interactions at quarter-filling in the strong coupling limit. We find two different charge-ordered ground states as the strength of the longer range interactions is varied. At lower energies, these CO states drive two different spin-ordered ground states. A variety of response functions computed here bear a remarkable resemblance to recent experimental observations for organic TMTSF systems, and so we propose that these systems are proximate to a QCP associated with $T = 0$ charge order. For a ladder system relevant to $\mathrm{Sr}_{14}\mathrm{Cu}_{24}\mathrm{O}_{41}$, we find in-chain CO, rung-dimer, and orbital antiferromagnetic ordered phases with varying interchain couplings and superconductivity with hole-doping.

Electron crystallization, or charge ordering (CO) due to interactions, is an issue of enduring interest in condensed matter physics. The study of the conditions favoring CO, along with its competition with metallic and/or superconducting states constitutes a problem of wide-ranging interest for a host of real systems \cite{1} composed of weakly coupled single chains/ladders.

In this work, we study this issue within an extended quarter-filled Hubbard model on a linear chain, described by,

$$
H_\text{eff} = -t \sum_{i,\sigma} (C_i^{\dagger} C_{i+1,\sigma} + h.c) + (U - 2zP) \sum_i n_{i\uparrow} n_{i\downarrow} + V \sum_i n_{i} n_{i+1} + P \sum_i n_{i} n_{i+2} \tag{1}
$$

In one-dimension, the spin fluctuations are those of an ideal $S = 1/2$ XXX AF chain, while the charge fluctuations are described by the Hamiltonian

$$
H_c = -t \sum_i (c_i^{\dagger} c_{i+1} + h.c) + (V - J/4) \sum_i n_{i} n_{i+1} + P \sum_i n_{i} n_{i+2} \tag{2}
$$

that describes a model with frustrating interactions. In 1D, the projected fermions are spinless fermions with a hard-core constraint. This model has been considered as a model for studying

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the effect of frustration on electron crystallization [2]. For narrow-band systems, we consider the limit $t << (V-J/4), P$. In this regime, we employ an extension of the trick used for the 1d next-nearest neighbor Ising chain: for $t = 0$, we notice that with $(V-J/4) > 2P$, the ground state is the usual CDW (Wigner) crystal for $n = 1$. With $2P > (V-J/4)$, however, the dimerized state (Peierls) is the ground state, written schematically as $(11001100...)$.

On the other hand, Peierls dimerization is possible because of the known exact solution of the 1D transverse field Ising model), and $(\cdots(01)(10)(01)(10)\cdots)$. Associating a pseudospin $\tau = 1/2$ operator, with $\tau^z = \pm 1$ for $(01)$ and $-1$ for $(10)$, the state is antiferromagnetic and doubly degenerate in terms of the $\tau^z_i$. For small $t$, this is an attractive trick because (in spin language) the transverse term does flip the $\tau^z_i$, but cannot break a pair. So one obtains,

$$H_{\text{eff}} = -\sum_l [2t\tau^x_l + (V-J/4 - 2P)\tau^z_l \tau^z_{l+1}]$$

This is just the Ising model in a transverse field, which has been studied extensively in 1D. If $(V-J/4-2P) < 0$, the ground state is ferromagnetically ordered in $\tau^z$, i.e., it corresponds to a Wigner CDW. For $(V-J/4-2P) > 0$, the Peierls dimer order results in the ground state. At $(V-J/4-2P) < 2t$, the quantum disordered phase has short-ranged pseudospin correlations, and is a charge “valence-bond” liquid. The quantum critical point at $(V-J/4-2P) = 2t$ separating these phases is a deconfined phase with gapless pseudospin $\langle \tau \rangle$ excitations, and power-law fall-off in the pseudospin-pseudospin correlation functions. Correspondingly, the density-density correlation function has a power-law singular behavior at low energy, with an exponent $\alpha = 1/4$ characteristic of the 2D Ising model at criticality. For $P = 0$, the metallic phase for $V \leq 2t$ is a Luttinger liquid, and in this limit, the low-energy physics is qualitatively similar to that of the usual $t-J$ model. The “Mott” insulating state for $V > 2t$ has Wigner CO in the ground state, and the M-I transition is of the Kosterlitz-Thouless type [3].

The full Hamiltonian in our case for the strong-coupling limit is now given by

$$H_{\text{eff}} = -\sum_l [2t\tau^x_l + (V-J/4 - 2P)\tau^z_l \tau^z_{l+1}] + J \sum_l S_l S_{l+1}$$

To study the magnetic phases, we adapt the Ogata-Shiba [4] technique for our case. This is possible if $J << t, V$, in which case, the pseudospin part is first solved exactly (this is possible because of the known exact solution of the 1D transverse field Ising model), and the exchange part is then treated as a perturbation. Writing the total wavefunction as a product of a spin and pseudospin wavefunction (where the spin wavefunction is defined in a Hilbert space of dimension $2^N$), i.e., $|\psi > = |\tau > \otimes |S >$, and following standard degenerate perturbation theory, the spin degeneracy is lifted by the correction (order $1/L$):

$$<H_{\text{eff}}>' = -2t <\tau^x> + \sum_l J_{l,l+1} (S_l S_{l+1} - 1/4)$$

where the average $<..>'$ denotes that the average is taken over the exact ground state $|\tau >$ of the pseudospin part above, i.e. $<A>' = <\tau |A|\tau >$ and $J_{l,l+1} = |V-J/4 - 2P| <\tau^z_l \tau^z_{l+1}>$. An interesting fact now emerges: Wigner CO (FM order of $\tau$) results in an HAFM $S = 1/2$ spin model with the Hamiltonian $H_s = J \sum_i S_i S_{i+1}$. This gives rise to a gapless AF ground state for the spin degrees of freedom. The charge (holon) excitations are gapped; this corresponds to a linear confining potential for holons. On the other hand, Peierls dimerization in the charge sector (AF Neel order of $\tau$) gives rise to dimerization in the spin sector, with the Hamiltonian $H_s = J \sum_i [1 + (-1)^i \delta S_i S_{i+1}]$.

Translated into fermion variables, this yields a sine-Gordon problem with $\beta^2 = 2\pi$, and describes an instability to a singlet pinned ground state commensurate with the Peierls CO
setting in at higher energies. The elementary excitations are solitons carrying $S^z = \pm 1$. Scaling theory predicts a dimer gap, $\Delta_d \approx \delta^{2/3}$. Exactly at $\beta^2 = 2\pi$, the SG model has just two $S^z = 0$ breather excitations with opposite parity [5], the lowest, even parity breather being degenerate with the $S^z = \pm 1$ soliton doublet, forming a $S = 1$ triplet, while the second odd-parity breather is a singlet with a gap, $\sqrt{3}\Delta_d$. It is important to notice that both charge and spin order arise from long range Coulomb interactions, and do not involve an electron phonon coupling mechanism.

Let us consider the implications of having the CO state in the high-$T$ regime, where one could imagine the system to be effectively one-dimensional. In particular, we want to look at the $\omega, T$ dependence of the various response functions at high-$T$. Using the exact solution of the pseudospin model in 1D, the high $T$ (in the “quantum critical” regime) behavior can be explicitly derived [6]. In fact, near Ising criticality, the response function, $\chi(r) \approx r^{-1/4}$ where $r = (x^2 + \tau^2)^{1/2}$ (with the velocity $v$ set to unity). This relation is still valid away from criticality in the “short range” regime, $r \ll \Delta_r$, where $\Delta_r$ is the pseudospin (in our case charge gap) gap of the 1D-TFIM. Using this asymptotic form, we have

$$\chi_{\text{crit}}(0, \omega) = -\frac{\sin(2\pi \Delta)}{(2\pi T)^{2-4\Delta}} B^2(\Delta - iS, 1 - 2\Delta)$$

(6)

where $S = \frac{\omega}{(2\pi T)^2}$, and $\Delta = 1/16$ is the conformal dimension. $B(x, y)$ is the beta function.

In the quantum critical region, an illuminating form is

$$\chi(k, \omega) = \frac{\chi(0, 0)}{1 - i\omega/\Gamma_R + k^2 \xi^2 - (\omega/\omega_1)^2}$$

(7)

where $\Gamma_R = (2\tan(\pi/16)) k_B T/h e^{\Delta_r/k_B T}$, $\omega_1 = 0.795(k_B T/h)$ and $\xi = 1.28(c\hbar/k_B T)e^\Delta_r/k_B T$, are determined solely by $T$ and the fundamental natural constants, as expected in the QC regime. Here, $\Delta_r$ is the energy gap to charge excitations in the Wigner/Peierls CO states described above. This represents the collective charge susceptibility, and the optical conductivity follows directly from $\sigma(\omega) = -\omega \chi(0, \omega)$, giving,

$$\sigma(\omega) = \frac{\chi(0, 0)}{\Gamma_R} \frac{\omega^2}{(1 - \omega^2/\omega_1^2)^2 + (\omega/\Gamma_R)^2}$$

(8)

The corresponding frequency-dependent dielectric function is obtained from $\epsilon(\omega) = 1 + (4\pi \sigma(\omega)/\omega)$, and the electronic contribution to the Raman scattering is estimated therefrom to be given by $I_R(\omega) = \text{Im}(1/\epsilon(0, \omega))$, for light polarized along the chain axis. In terms of the charge susceptibility, this is simply,

$$I_R(\omega) = \text{Im} \frac{1}{\epsilon(\omega)} = \frac{4\pi \chi(0, 0)}{\Gamma_R} \frac{F(\omega, T)}{1 + (4\pi \chi(0, 0)/\Gamma_R)^2 F^2(\omega, T)}$$

(9)

where $F(\omega, T) = \frac{\omega}{(1 - \omega^2/\omega_1^2)^2 + (\omega/\Gamma_R)^2}$.

$\chi''(k, \omega)$ has its maximum value at $\omega_m = \omega_1 - \omega/\Gamma_R$, implying that the collective mode broadens and shifts to higher energy linearly in $T$ with increasing $T$ at high temperatures. Further, the $T$-dependent damping rate of the collective mode correlates well with the relaxation peak seen in transport, underlying their common origin. In fact, the $dc$ resistivity is linear in $T$ at high $T$, with “insulating” features showing up at lower $T$. In our picture, these are collective (longitudinal) bosonic charge-density modes in the high-$T$ quantum critical region above an incipient CO transition (expected to occur at low $T$). In fig.(1), we show the
Fig. 1 – A three-dimensional plot of the Raman intensity $I_R$ versus a scaled energy $\omega/T$ and temperature $T$ for parameter values of the original model which constitute a gap $\Delta_\tau = 0.05k_B$.

The electronic Raman lineshape as a function of $\omega/T$. The sharp low energy peak corresponds to the collective charge density fluctuation mode of the CO ground state.

The corresponding frequency-dependent dielectric constant also shows an explicit $\omega/T$ scaling in the QC regime, or generally, at high-$T$, it shows strong $T$-dependence. From fig.(2), we see that it becomes $\omega$-independent at high $T$, but appreciably increases as $T$ is lowered, with a maximum at $\omega \simeq T$.

Fig. 2 – A three-dimensional plot of $\sigma(\omega)/\omega$ versus a scaled energy $\omega/T$ and temperature $T$ for $\Delta_\tau = 0.05k_B$. The limiting behaviors are: (1) $\sigma(\omega) \simeq \frac{\omega^2}{T}$ for high $T$, and $\sigma(\omega) \simeq \frac{\omega^3}{T}$ for low $T$. (2) $\epsilon'(\omega, T) = 1 + \frac{T_{11}}{T}$ for high $T$, and $\epsilon'(\omega, T) \simeq -\frac{T_{11}}{\omega}$ for low $T$.

The fact that organic charge transfer salts [7] exhibit features very similar to those found above has interesting implications. In light of our results, these anomalous features can now be identified with proximity to an underlying quantum critical point associated with charge (Wigner/Peierls) ordering. We recall that very recent work [8, 9] shows that the dimerized insulating state in TMTSF systems has charge order at low $T$. Interestingly, $\epsilon'(0, \omega)$ indeed
shows appreciable increase as $T$ is lowered, further supporting an interpretation based on proximity to an underlying CO ground state [7]. Hence, we conclude that observation of these features in TMTSF systems constitute strong evidence that this system is close to a putative QCP associated with charge order. Observation of dimerized/Neel ordered AFM states co-existing at low $T$ are also naturally understood in light of the analysis above [9].

Two-chain Ladders We now consider the strong coupling version of a coupled two-chain ladder system, with each chain being described by $H$ as in eq.(1). In the strong coupling limit, where each chain is described by a TFIM for charge degrees of freedom, the coupled chain model is constructed as follows. For $U \rightarrow \infty$, and $V,P > t$ (but $(V - 2P)$ comparable to $t$), the charge degrees of freedom of the fermionic problem for each chain are described by an effective pseudospin model on n-n bonds, via the effective Hamiltonian,

$$H^{\text{chain}} = -\sum_j [2t \sigma^x_j + (V - J/4 - 2P) \sigma^z_j \sigma^z_{j+1}]$$

(10)

Rotating the pseudospin axis such that $\sigma^x \rightarrow \sigma^z$, $\sigma^z \rightarrow -\sigma^x$ and coupling two such chains via an interaction coupling $U_\perp$ and a two-electron interchain transfer $t_\perp$, we have the effective Hamiltonian for the charge sector of the two chain system as

$$H = -\sum_{j,a} [2t \sigma^z_{j,a} + (V - J/4 - 2P) \sigma^z_{j,a} \sigma^z_{j+1,a}] - \sum_{j,a,b \neq a} [U_\perp \sigma^z_{j,a} \sigma^z_{j,b} + t_\perp (\sigma^x_{j,a} \sigma^x_{j,b} + \sigma^y_{j,a} \sigma^y_{j,b})]$$

(11)

where $a,b = 1,2$ is the chain index. Denote the in-chain pseudospin coupling as $J = (V - J/4 - 2P)$ and the inter-chain pseudospin coupling as $J_\perp = U_\perp$. Here, we study the strong coupling version of this problem in two limits (see below). The weak-coupling problem is studied elsewhere [10].

For the case of $|J_\perp| \gg |J|$, $t_\perp$, the 2 chain system can be better thought of as strongly-coupled chains which are weakly coupled to their neighboring rungs. Thus, we treat $J$ as a weak perturbation on the zeroth-order system of rungs defined by the large coupling $J_\perp$, giving $H_{\text{eff}} = H_0 + H_1$ where

$$H_0 = -\hbar \sum_{j,a} \tau^z_{j,a} + J_\perp \sum_{j,a,b \neq a} \tau^z_{j,a} \tau^z_{j,b}$$

$$H_1 = -J \sum_{j,a} \tau^x_{j,a} \tau^+_{j+1,a} - \frac{t_\perp}{2} \sum_{j,a,b \neq a} (\tau^+_{j,a} \tau^-_{j,b} + \text{h.c})$$

(12)

where the effective magnetic field is given by $h = 2t > 0$.

LEH for $J_\perp < 0$. For $J_\perp < 0$ and $h \ll J_\perp$, we find that the triplet state $|+\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$ and the singlet state $|-\rangle = \frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle - |\downarrow\downarrow\rangle)$ are degenerate on any rung and are separated from all other states by a large gap of order $J_\perp$. Thus, these two states define the subspace which will determine the low-energy physics of the system. Identifying a pseudospin-1/2 operator $\xi_j$ with the low-energy subspace on each rung, we treat the Hamiltonian $H_1$ as a perturbation (to second order in $J/J_\perp$) to obtain the LEH as

$$H = \sum_j \left(-\frac{J^2}{2J_\perp} \left(\frac{J^2}{J_\perp^2} - \frac{2h^2}{J_\perp^2} \right) \xi^z_j \xi^z_{j+1} - \frac{t_\perp}{2} \sum_j \xi^z_j \right)$$

$$-\frac{J^2}{2J_\perp} \left(\frac{h^2}{J_\perp^2} \right) \left(\xi^z_j \xi^+_{j+1} + \text{h.c} \right) - \frac{J^2}{8J_\perp} \left(\frac{J^2}{J_\perp^2} - \frac{2h^2}{J_\perp^2} \right)$$

(13)
We find that \( t_\perp \) acts as the strength of a Zeeman-splitting like term in the LEH. Bosonising this, we obtain a sine-Gordon Hamiltonian with a cosine potential in the dual (\( \theta \)) field and a magnetic-field term
\[
H = \frac{\nu}{2}[(\partial_x \phi)^2 + (\partial_x \theta)^2] - \frac{m}{2\pi \alpha} \cos \beta_1 \theta - \frac{\beta_1 t_\perp}{2} \partial_x \phi
\] (14)

We note that bosonisation of the general XYZ Hamiltonian results in the appearance of an additional \( 4k_F \) Umklapp term \([11]\), \( \cos \beta_2 \phi \), which is irrelevant for a finite \( t_\perp \) and is hence ignored in what follows. When \( t_\perp \) is below a certain critical value, incommensurate Wigner charge order (ordering of the \( \xi_z \)) occurs \([12]\). Above this critical value, a spin-flop transition orders the system in the \( x \) direction (i.e., ordering of the \( \xi_x \)) via a Kosterlitz-Thouless transition. For \( \beta_2^2 < 8\pi \), the cosine in the dual field is a relevant perturbation and orders the dual field. The magnetic-field term \( \propto t_\perp \) leads to a ground state with charges which are coherently delocalised on the diagonals of each pair of nearest-neighbor rungs; this is an orbital antiferromagnet-type ordering with circulating currents in plaquettes \([3, 11]\).

**LEH for \( J_\perp > 0 \)**. For \( J_\perp > 0 \), and \( h > 0 \), the triplet state \(|+\rangle = |\uparrow\uparrow\rangle\) is the low energy state on any rung. For \( h = 0 \), we find that the triplet states \(|+\rangle \) (defined above) and \(|-\rangle = |\downarrow\downarrow\rangle\) are degenerate. Thus, we can again identify these two states as the subspace which determines the low-energy physics of the system. For \( h \ll J \), we again identify a pseudospin-1/2 operator \( \xi_j \) with the low-energy subspace on each rung, and treat the Hamiltonian \( H_1 \) as a perturbation (to second order in \( J/J_\perp \)) to obtain the LEH as
\[
H = -\frac{J^2}{4J_\perp} \sum_j \xi_j^x \xi_{j+1}^x - 2t \sum_j \xi_j^z .
\] (15)

This is just the 1D TFIM (with ferromagnetic Ising coupling). In the ordered phase, the ground state has in-chain Wigner CO and dimers on every alternate rung. The disordered phase is a gapped, short-ranged charge-dimer liquid. At \( t = J^2/4J_\perp \), the quantum critical point describes a gapless charge-dimer liquid with \( \omega/T, \nu k/T \) QC scaling, exactly as was described before. Transposing the results obtained before, we conclude that the dc resistivity, optical conductivity, electronic Raman and dielectric responses will be exactly described by the same scaling functions (eqs.\((6)-(9)) \) with the gap, \( \Delta_r \), now being the CO gap of the ladder problem (\( H \) in eq.\((15))\). Very interestingly, exactly such behavior is observed in undoped ladder system \( Sr_{14}Cu_{24}O_{41} \) \([13]\) and attributed to a longitudinal, collective charge fluctuation mode, exactly as described here.

**LEH for hole-doped ladder.** Upon doping the ladder with holes, while a single hole experiences a linear confining potential in the Wigner (Ising-like) or Peierls (dimerized) CO background, a pair of holes on the same rung is free to propagate. One can then describe the hole-pair as a hard-core boson, representing its creation and annihilation operators using the spin-1/2 operators \( \sigma^\pm \); the local charge density is then described by \( \sigma^z \). Following \([3]\), we find the LEH describing the dynamics of such hole-pairs to be the XXZ model in an external magnetic field
\[
H = \sum_j \left[ -\frac{t_h}{2}(\sigma_j^+ \sigma_{j+1}^+ + h.c) - u_h \sigma_j^z \sigma_{j+1}^z - \mu \sigma_j^z \right]
\] (16)

where \( t_h \sim J^2/J_\perp \) is the pair-hopping matrix element, \( u_h \) is the Coulomb interaction between pairs on nearest-neighbour rungs and \( \mu \) is the chemical potential of the holes. The phase diagram of this model is known \([3]\); for \( \mu = 0 \) and \( u_h > t_h \), the ground state is an insulating
CDW of hole pairs. Beyond a critical $\mu_c = f(u_h, t_h)$, the system has a ground state described by Bose condensation of hole pairs. In fact, from the bosonisation analysis of the equivalent $S = 1/2$ XXZ model in an external Zeeman field, we know that $<\sigma_i^z \sigma_i^{z+r}> \simeq r^{-1/\alpha}$ and $<\sigma_i^x \sigma_i^{x+r}> \simeq r^{-\alpha}$ where $\alpha = 1/2 - \pi^{-1} \sin^{-1}(2u_h/t_h)$. Clearly, for $\alpha < 1$, the ground state has dominant superconducting correlations. This is true for both the cases described above: in the first case, we have a Bose condensate of intrachain pairs of holes, while in the second hole pairs on individual rungs Bose condense, describing two possible superconducting types in the ladder system. This finding matches our conclusions obtained from a weak coupling analysis [10], and thus constitutes a generic feature of undoped/doped strongly correlated ladder systems. 

To conclude, we have explored the strong-coupling limit of strongly correlated single chain and two-leg ladder models using a variety of methods. Our results strongly suggest that prototype examples like organics (TMTSF) and $Sr_{14}Cu_{24}O_{41}$ lie in close proximity to an underlying QCP associated with charge order, and constitutes an advance in our understanding of their physical responses in a new theoretical framework.

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REFERENCES

[1] McCarron III E. M. et al., Mater. Res. Bull., 23 (1988) 1355; Uehara M. et al., J. Phys. Soc. Jpn., 65 (1996) 2764; Jerome D., Organic Superconductors: From (TMTSF)$_2$PF$_6$ to Fullerenes (Marcel Dekker, New York) 1994, p. 405.

[2] Zhuravlev A. K. and Katsnelson M. I., Phys. Rev. B, 64 (2001) 033102.

[3] Gogolin A. O., Nersesyan A. A. and Tsvelik A. M., Bosonization and Strongly Correlated Systems (Cambridge University Press, Cambridge) 1998 and references therein.

[4] Ogata M. and Shiba H., Phys. Rev. B, 41 (1990) 2326.

[5] Haldane F. D. M., Phys. Rev. B, 25 (1982) 4925.

[6] Sachdev S., Quantum Phase Transitions (Cambridge University Press, Cambridge) 1999 and references therein.

[7] Brazovskii S., cond-mat/0401309 preprint, (2004) and references therein; Staresinic D. et al., cond-mat/0509146 preprint, (2005).

[8] Brazovskii S., Monceau P. and Nad F., Synthetic Materials, 137 (2003) 1331.

[9] Fehske H. et al., Physica B, 359-361 (2005) 699.

[10] Lal S. and Laad M. S., to be submitted, (2005).

[11] Giamarchi T., Quantum Physics in One Dimension (Oxford University Press, Oxford) 2004 and references therein.

[12] Giamarchi T. and Schulz H. J., J. de Physique (Paris), 49 (1988) 819.

[13] Blumberg G. et al., Science, 297 (2002) 584.