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Incipient quantum melting of the one-dimensional Wigner lattice

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Abstract

A one-dimensional tight-binding model of electrons with long-range Coulomb interactions is studied in the limit where double site occupancy is forbidden and the Coulomb coupling strength V is large with respect to the hopping amplitude t. The quantum problem of a kink–antikink pair generated in the Wigner lattice (the classical ground state for $t = 0$) is solved for fillings $n = 1/s$, where s is an integer larger than 1. The pair energy becomes negative for a relatively high value of V, $V_c/t \approx s^3$. This signals the initial stage of the quantum melting of the Wigner lattice.

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1. Introduction

Three decades ago, Rice et al. have introduced nonlinear phase excitations of a charge-density wave condensate as a new type of charged states and referred to them as φ-particles [1]. Their arguments were based on the peierls instability of a one-dimensional coupled electron–phonon system, in the limit where the charge-density wave amplitude is much smaller than the average electronic charge-density. Shortly after, Hubbard discussed qualitatively the role of dimer pairs (domain boundaries) in a nearly quarter-filled band with dominant long-range Coulomb interactions [2]. As we will argue, the two types of charged states are closely related.

We consider a one-dimensional system of spinless fermions described by the Hamiltonian

$$H = -t\sum_{t} (c_i^\dagger c_{i+1} + c_{i+1}^\dagger c_i) + \sum_i \sum_{l\geq 1} V_l n_i n_{i+l}, \qquad (1)$$

where $c_i^\dagger$ and c_i are fermionic creation and annihilation operators, respectively, $n_i = c_i^\dagger c_i$, c is the occupation number of site i and $V_l = V/l$ represents the long-range Coulomb potential. Such a model may describe molecular chain compounds where the interaction between two (valence) electrons on the same molecule is so large that double occupancy can safely be discarded. The spin quantum number is then redundant because the exchange of two electrons is dynamically forbidden.

We have in mind systems where the average site occupation is a rational number between 0 and 1, $n = r/s$. Depending on the band filling the fermions then represent either electrons or holes. The classical ground state ($t = 0$) is charge-ordered and forms a "generalized Wigner lattice" [2]. In the special case $n = 1/s$, the unit cell contains s sites, one of which is occupied. A generalized Wigner lattice is clearly insulating and therefore the long-range Coulomb interactions remain unscreened. Insulating charge-ordered phases have been reported for organic chain compounds. For a collection of recent results see [3][1] and in several cases the nature of charge ordering is more likely that of a Wigner crystal than that of a (small-amplitude) charge-density wave.

We have recently used a variational wave function for describing the modifications of the classical ground state (the generalized Wigner lattice) induced by a small but finite hopping term ($t \ll V$) [4]. Here we consider the same regime, but follow a different route by introducing the notion of charge defects, as was done in [5] in the case of

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[1] More references can be found in [4], together with a discussion on the possible relevance of the long-range interactions in specific quasi one-dimensional materials.

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electron–phonon interactions. Such defects occur in pairs (kinks–antikinks) and can be viewed as fractionally charged particles, in close analogy to the φ-particles of Michael Rice.

2. Kink–antikink pair for $n = 1/2$

We start our discussion for a density $n = 1/2$. In order to guarantee overall charge neutrality we introduce a rigid compensating background ($n = 1/2$ charge of opposite sign at each site). The classical ground state in this case corresponds to alternating filled and empty sites (Fig. 1a). Note that this configuration, already at the classical level, is doubly degenerate, since exchanging the empty and occupied sites does not change the overall energy. The most simple defect in this perfect structure, called kink in the following, is a domain boundary separating the two possible ground state configurations. It can be visualized as a pair of occupied nearest-neighbor sites (a dimer) and is necessarily accompanied by an antikink, a pair of empty nearest-neighbor sites.

The classical energy of a kink–antikink pair is readily evaluated. Introducing a pair of size $2d$, i.e. a kink and an antikink separated by a distance $2d$ (see Fig. 1), costs an energy

$$\Delta(d) = \sum_{p=1}^{d}\sum_{m=p}^{\infty}[V_{2m+1} + V_{2m-1} - 2V_{2m}]. \tag{2}$$

This sum can be transformed into a power series in $1/2d$ with leading terms

$$\Delta(d) = \frac{V}{2} - \frac{V}{4}\frac{1}{2d} + \frac{V}{8}\frac{1}{(2d)^3}. \tag{3}$$

Here the first term represents the creation energy of two well separated defects. The second term is the Coulomb attraction between kink and antikink with a coupling reduced by a factor of four as compared to the original Hamiltonian (1). Therefore, the defects carry effective charges $\pm 1/2$ (measured in units of the electronic charge). This result agrees with what one obtains by adding or subtracting an electron to the classical ground state configuration [2]. In fact, adding an electron at an empty site results in a high-energy configuration with a three-electron cluster. The energy is lowered

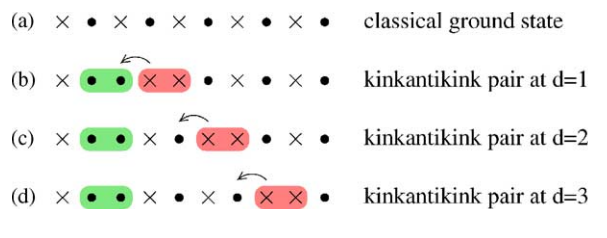

Fig. 1. Classical ground state and low-energy defect pairs at $n = 1/2$. Dots/crosses stand for filled/empty sites respectively. The kinks and antikinks correspond to the shaded areas. The arrows indicate the hopping processes leading to a given configuration.

by moving away one of these electrons, thus creating a pair of defects (two kinks in our language). Since the overall added charge is -1, each kink carries a charge $-1/2$. Similarly, removing an electron produces two antikinks, each of which carries a charge $+1/2$. The third term in Eq. (3) can be interpreted as the interaction of two dipole moments pointing in opposite directions parallel to the chain axis. The size of the dipoles is equal to the fractional charge times half a lattice constant, in our units 1/4. We conclude that kinks and antikinks behave like fractionally charged particles with electric dipole moments. The dominant interaction is the Coulomb attraction, even at the shortest possible distance $2d = 2$.

We now turn to the quantum problem of a defect pair, i.e. we diagonalize the Hamiltonian within the subspace of states $|m, d\rangle$, corresponding to a kink at site $m - d$ and an antikink at site $m + d$ (m thus indicates the center-of-mass of the pair). In general the effect of a hopping event on the state $|m, d\rangle$ is either to move an existing defect by two lattice sites, which modifies both the size ($2d \to 2d \pm 2$) and the center ($m \to m = \pm 1$) of the pair (see Fig. 1), or to create (annihilate) an additional pair. Restricting ourselves to the subspace of single-pair states, creation, and annihilation processes are forbidden and the quantum problem reduces to the eigenvalue equation

$$\begin{aligned}&[H - \Delta(d)]|m, d\rangle t + [|m-1, d+1\rangle + |m+1, d-1\rangle \\ &\quad + |m+1, d+1\rangle + |m-1, d-1\rangle] = 0\end{aligned} \tag{4}$$

The center-of-mass and relative motions can be separated by introducing the Bloch superposition

$$|\psi\rangle = \sum_{m,d\geq 1} e^{iKm}\psi(d)|m, d\rangle. \tag{5}$$

Introducing this Ansatz into Eq. (4) leads to the following eigenvalue equation for the wave function $\psi(d)$,

$$\begin{aligned}&E\psi(d) = \Delta(d)\psi(d) - 2t\cos K[\psi(d+1) + \psi(d-1)],\\ &\quad d \geq 1.\end{aligned} \tag{6}$$

Seeking for the lowest-energy excited state we restrict ourselves to the pair at rest ($K = 0$). The classical ground state state (no defect pairs) is excluded by imposing the boundary condition $\psi(0) = 0$.[2] This eigenvalue problem can be treated to arbitrary accuracy numerically, but more insight is gained by solving it in the continuum limit, $\psi(d) \to \psi(x)$, $\psi(d\pm 1) \to \psi(x\pm 2)$,where x is the position in units of the distance between neighboring sites. Neglecting the dipolar interaction in Eq. (3) we obtain the eigenvalue equation

$$\varepsilon\psi(x) = -8t\frac{d^2\psi}{dx^2} - \frac{V}{4|x|}\psi(x), \tag{7}$$

[2] We seek an excited state which is orthogonal to the classical ground state. This is the natural procedure for estimating at which point in parameter space a trial ground state becomes the “wrong vacuum.”

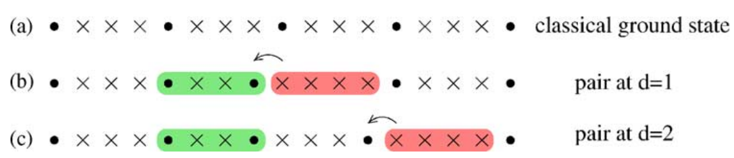

Fig. 2. Classical ground state and low-energy defect pairs at $n = 1/4$. The symbols are the same as in Fig. 1.

where $\varepsilon = E - V/2 + 4t$. Eq. (7) is equal to the radial eigenvalue equation of the hydrogen problem in the s-wave channel, and the boundary conditions are also the same, $\psi(0) = 0$. Therefore, we can immediately write down the lowest-energy wave function of a pair,

$$\psi_0(x) = 2a_0^{-3/2} x \mathrm{e}^{-x/a_0}, \quad x \geq 0, \tag{8}$$

with an effective Bohr radius

$$a_0 = \frac{64t}{V} \tag{9}$$

and an energy

$$E_0 = -4t + \frac{V}{2} - \frac{V^2}{512t}. \tag{10}$$

When the interaction strength is lower than the critical value $V_c/t = 8.27$, $E_0 < 0$ and the formation of a kink–antikink pair becomes favorable by virtue of the gain in energy achieved through delocalization as a Bloch state.[3]

When approaching the critical interaction strength, the classical Wigner lattice will be destabilized due to a proliferation of kink–antikink pairs. To study this problem it would be necessary to take into account the dynamically screened interaction between defects, a task beyond the scope of this work. However, we do not expect a true transition to a one-dimensional metallic state (a Luttinger liquid), because for arbitrarily small V the system is unstable with respect to a charge-density wave with the same period as for the Wigner lattice [6]. Therefore the critical strength V_c indicates a crossover between a Wigner lattice and small-amplitude charge-density wave. Note that in the crossover region the binding energy $V^2/512t$ makes a negligible correction to the quantum mechanical energy of a pair: the main contributions to Eq. (10) are the delocalization energy $-4t$, and the formation energy of the isolated defects $V/2$.

3. Kink–antikink pairs for *n=1/s*

We extend now the considerations of the previous section to other filling factors $n \neq 1/2$. For a general value of n already the classical limit ($t = 0$) is quite involved, leading to complicated charge patterns [2]. Therefore it is worthwhile to make contact to the opposite limit, $V < t$, where a simple picture is available. Mean-field theory applied to the Hamiltonian (1) yields a ground state with a modulated charge-density

$$\langle \delta n_l \rangle = a \cos(Ql + \varphi_l), \tag{11}$$

where $Q = 2k_F = 2\pi n$ in the case of spinless fermions, and $a \ll n$ for $V < t$. The phase φ_l is an arbitrary constant for an incommensurate situation (n irrational), but locked to one of s possible values in the commensurate case, where $n = r/s$ with integer numbers r and s [7]. Adding an electron to an incommensurate charge-density wave results in a slight shift of the wave vector Q, $Q \to [1 + (1/L)]Q$, where L is the chain length. In view of Eq. (11), this effect can also be attributed to a phase shift $\varphi_l = 2\pi l/L$. For a commensurate case the phase achieves the change of 2π from $l = 0$ to L through s steps, i.e. there are s φ-particles, each of which carries a fractional charge $-1/s$.

Let us now return to the large V limit and restrict ourselves to simple ratios $n = 1/s$, $s \geq 3$. In this case the classical ground state is s-fold degenerate and there are several types of domain walls separating the different configurations. It turns out that the low-energy domain walls are those which connect nearby ground state configurations, i.e. Wigner lattices where the locations of electrons differ by one lattice constant (see Fig. 2). These "kink" and "antikink" defects again occur in pairs. Repeating the arguments of the steps of the previous section, we evaluate the classical energy of a kink–antikink pair of size sd,

$$\Delta(d) = \sum_{p=1}^{d} \sum_{m=p}^{\infty} (V_{sm+1} + V_{sm-1} - 2V_{sm}). \tag{12}$$

The summation can again be performed for large sizes, and we find up to first order in $1/sd$

$$\Delta(d) = \frac{V}{s}\left[1 - \frac{\pi}{s}\cot\left(\frac{\pi}{s}\right)\right] - \frac{V}{s^2}\frac{1}{sd}. \tag{13}$$

The first term, the pair creation energy Δ_∞ at infinite separation, agrees with the corresponding quantity in Eq. (3) for $s = 2$. The second term corresponds to the Coulomb attraction for two particles with fractional charges $\pm 1/s$ at a distance sd, in agreement with the counting argument presented above for the φ-particles.

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[3] The continuum description is appropriate provided that the extent $\langle x \rangle = 96t/V$ is larger than the average distance between particles, a condition that is well satisfied at the critical coupling V_c. Indeed, solving the original discrete problem of Eq. (6) numerically yields $V_c/t = 8.25$, which is very close to the continuum estimate.

We can again use the continuum limit for calculating the lowest-energy quantum pair state for general s. The wave function is again given by Eq. (8), but with a Bohr radius

$$a_0 = \frac{4s^4 t}{V}. \tag{14}$$

Thus the pair size increases strongly with decreasing density $n = 1/s$. Correspondingly, the binding energy decreases, as seen in the pair energy

$$E_0 = -4t + \Delta_\infty - \frac{V^2}{8s^6 t}, \tag{15}$$

where the first two terms dominate for large s. In fact, for $s \gg 1$ we can safely use the asymptotic value

$$E_0 \approx -4t + \frac{\pi^2}{3s^2} V \tag{16}$$

to locate the crossover region, where this energy becomes negative. The result,

$$\frac{V_\mathrm{c}}{t} \approx \frac{12}{\pi^2 n^3} \tag{17}$$

agrees well with our previous variational estimate [4]. This is not surprising because the creation of a pair can be achieved through a hopping event that moves a particle out of the ground state configuration. The probability of such a hopping event was evaluated in [4], and was used to determine a criterion for the instability of the generalized Wigner lattice.

In contrast to the case $n = 1/2$, for lower density this criterion does not signal a crossover to a small-amplitude charge-density wave, it rather indicates that part of the electronic charge is spilled over to neighboring sites of the classical Wigner lattice.

4. Discussion

In this paper we have determined the lowest-energy quantum state of a kink–antikink pair in a one-dimensional generalized Wigner lattice. We have calculated the critical value V_c of the interaction strength below which the pair energy is negative, and charge defects will be generated spontaneously. Kinks (or antikinks) are the strong-coupling analogs of the φ-particles studied a long time ago by Rice et al. [1].

The true quantum ground state of electrons interacting through long-range Coulomb forces contains kink–antikink pairs due to quantum fluctuations, even above V_c. It would be interesting to proceed from the single-pair solution to that of an arbitrary number of pairs. This step is highly non-trivial, although at first sight it looks similar to that from the Cooper problem to the BCS wave function. One of the difficulties arises from the mutual interactions between pairs, another from their non-local character.

If the density of pairs in the ground state is large enough, they lose their identity, and kinks and antikinks may move rather independently. In this case one can imagine a dc charge transport due to moving defects. Whether such a mechanism is responsible for the observed Drude peak in the Bechgaard salts [8] is an interesting open question.

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