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Thermodynamics of a one-dimensional lattice system with long-range interelectron repulsion

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The thermodynamic characteristics of one-dimensional lattice electron systems with long-range interelectron interactions are explicated and studied at arbitrary temperature T and pressure P. Only interactions between neighboring particles are taken into account. Electron-electron correlations are found to be controlled only by the ratio P/T, increases in which produce a continuous transition from a weakly nonideal gas to an electron ''crystal'' with filling factor $1/q, q=1,2,\ldots$. This is a remnant of the devil's staircase which obtains at $T=O$. It is shown that with a decrease in P the crystal periods, q, increase, the transitions occurring via narrow regions of a liquidlike state. Charge carriers in the ''crystals'' are discrete solitons with a *fractional* charge $e^{*}=\pm e/q$. [S0163-1829(96)05631-7]

I. INTRODUCTION

Narrow-band conductors with long-range interelectron repulsions (LRIR's) have peculiar and very interesting properties that not only differentiate them qualitatively from metals (including the transition metals) and semiconductors but from the conductors of the Hubbard type, which are characterized by the *locality* of their electron-electron interaction.[1–3] The study of LRIR conductors seems to be particularly important and timely because layered structure high temperature superconductive metal oxides (even at ''metallic'' electron densities) and a number of metal oxides of other types are governed by LRIR. In addition, some change-transfer salts and artificial conductive systems (superlattices) belong to this class of materials.

As was recently shown by one of the authors (A.A.S.), the remarkable feature of narrow-band conductors with LRIR's is that inherent to them is a specific localized electron macroscopic state that is different from a Wigner crystal the current theory of the FEP is given in Ref. 4. This state we call the ''frozen'' electron phase (FEP). This arises from LRIR's in combination with the *discreteness* of the narrow-band electron dynamics (i.e., the narrow-band electrons move over the conductor by hopping between nearest-neighbor conductor-lattice sites with equivalent atomic orbitals) and exists over a wide parameter range determined by the following criterion:

$$t/U \leqslant C \sim 1. \tag{1}$$

Here t is the bandwidth, and $U\sim(a/R)E$ (a is the crystal spacing, R is the average distance between electrons, and E is the Coulomb energy per electron) is typical of the LRIR energy variation on electron hopping. In such a situation the complete destruction of the narrow-band Bloch states occurs, and electrons become localized within quantum traps of an atomic size. Due to the dynamic origin of this localization, heating of the system cannot release the electrons from their traps, and hence the FEP, unlike a Wigner crystal,[6,7] can not turn into Fermi liquid at any temperature. Another fundamental difference between the FEP and a Wigner crystal is that even at zero temperature a FEP can be in an ''electron glass'' state over all of parameter space (1). The electron glass state results from the fact that the ground state of an LRIR system with $t=0$ generally has a highly *disordered* (incommensurable) space structure at *a given electron density*. This incommensurability was discovered by Hubbard[1] in the one-dimensional (1D) case. In Ref. 5 it was pointed out that there is strong reason to believe that the disorder exists irrespective of the dimensionality. In the general case ($t\neq 0$) the electron glass is similar to a spin glass in many respects (nonergodic behavior, an infinite spectrum of relaxation times caused by infinite degeneration of the ground state, etc).[4]

Because of the above properties the FEP seems a very interesting subject for investigation. This is especially true at a finite temperature T, or in situations where the ratio t/U of criterion (1), is not too small. For example, as was shown in Ref. 5, a layered s-d system (which one can consider as a cuprate model) inevitably exhibits high-temperature superconductivity (with $T_c\sim t$) if the narrow-band subsystem (the d electrons) is in the *FEP electron glass* state. In spite of some advances in our understanding the nature of the FEP (see Ref. 4) there is still a great deal to learn to develop a consistent theory of its thermodynamics and kinetics. It seems appropriate to advance these studies by considering the *thermodynamics* of the LRIR systems, neglecting all dynamical effects produced by the finiteness of the bandwidth (i.e., $t=0$). Such an approach is applicable not only to narrow-band electron conductors but also to adatoms systems (if the adatom interaction with a substrate is sufficiently strong[8]) and other physical systems which exhibit similar behavior [for example, charge-transfer salts of a tetrathioflualene-tetracyanoquinodimethane (TTF-TCNQ) type]. The general thermodynamic problem appears to be rather complicated. Furthermore, as far as we know, the thermodynamics of 1D LRIR models has not yet been fully explicated, despite its appeal and relative simplicity. The main objective of our present study is to fill this gap in part.

II. APPROXIMATION

In accord with the above, we assume the Hamiltonian of the system under consideration to be of the form

$$\mathcal{H}=\mathcal{H}(l_1,l_2,\ldots,l_N)=\tfrac{1}{2}\sum_{\substack{m,n=1\\ m\neq n}}^{N}\varepsilon(l_m-l_n),\tag{2}$$

where the discrete independent variable l_m, ($m=1,\ldots,N$) is an mth electron coordinate measured in lattice units, N is the total number of electrons, and $\varepsilon(x)$ is the LRIR pair potential. This potential is a convex function of the continuous argument x which diminishes faster than x^{-1} but is otherwise arbitrary. To begin, it is necessary to investigate the properties of the ground state of the Hamiltonian (2). These were first considered by Hubbard,[1] who offered a rigorous procedure for constructing the ground-state configuration, which he aptly called the generalized Wigner lattice (GWL). As was later shown in Ref. 2, the Hubbard algorithm can be expressed by the simple formula

$$l_i=\left[i\frac{1}{\nu}+\phi\right].\tag{3}$$

Here $\nu=N/N_0$ is the electron density (filling factor), N_0 is the total number of the lattice sites (N_0 is the length L of the 1D system measured in lattice units), $1/\nu$ is the average interelectron distance; [ ] denotes an integer part, and ϕ is an arbitrary number. In what follows we study the usual thermodynamic limit (i.e., $N,N_0\to\infty$ in such a way that ν remains finite).

The distinctive feature of the GWL is that at given chemical potential, μ, only *rational* values of $\nu=p/q$ (p and q are integers) survive. That is, the GWL has a periodic electron lattice structure with p particles per cell and a period length q. This fact leads to a rather peculiar dependence of ν on μ, the so-called ''devil's staircase.'' This is a well-developed fractal structure[1,3,2] in which for each rational value of $\nu=p/q$ there is a finite interval Δ of the chemical potential μ within which ν is not changed. Following the reasoning of Bak and Bruinsma,[3] we find that

$$\Delta\mu=q\Delta\varepsilon(q),\tag{4}$$

where

$$\Delta\varepsilon(q)=\sum_{k=1}^{\infty}k[\varepsilon(qk+1)-2\varepsilon(qk)+\varepsilon(qk-1)].\tag{5}$$

The left $[\mu_d(p/q)]$ and right $[\mu_i(p/q)]$ endpoints of the $\nu(\mu)$ devil's staircase intervals are the changes of the ground-state energy resulting from decreasing and increasing N by *one particle*, respectively. The ground state and its space structure here described are of a universal nature, since they occur for all physically reasonable pair potentials $\varepsilon(x)>0$.

As will be seen from the following, the most convenient thermodynamic variables for our study are not T and μ, but instead T and the pressure P. The dependence of ν on P at $T=0$ is also a devil's staircase which differs from $\nu(\mu)$ only in the width of the step, ΔP. The endpoints of this devil's staircase, $P_d(p/q)$ and $P_i(p/q)$, are changes of the ground-state energy resulting from decreasing or increasing L, respectively, by *one spacing* at fixed N. (P is measured in energetic units, $P_i<P_d$.) Taking into account this fact, one can obtain the simple formula

$$\Delta P=p\Delta\mu/q=p\Delta\varepsilon(q),\tag{6}$$

as shown in the Appendix.

One should stress a remarkable feature of expressions (4) and (6), namely, that the devil's staircase intervals $\Delta\mu$ and ΔP are proportional to the value $\Delta\varepsilon$ (5), which is itself determined by the electron-electron interaction at distances $\gtrsim q$ *irrespective of the number of electrons*, p, *per unit cell.* Thus, $\Delta\mu$ and ΔP tend to zero as $q\to\infty$ according to $\partial^2\varepsilon(x)/\partial x^2|_{x=q}$. Hence the more ''irrational'' is the ratio p/q, the less is the lengths of the intervals (4) and (6). This suggests that at finite temperatures *all* GWL space structures of period q, fulfilling the condition $\varepsilon(q)\gg T$, are slightly perturbed by thermal fluctuations. Conversely, if q satisfies the condition $\varepsilon(q)\lesssim T$ the corresponding GWL structure will be essentially transformed or completely destroyed, smoothing out the devil's staircase intervals $\Delta P(q)$ and $\Delta\mu(q)$.

We will study the most realistic case, that of a rapidly declining pair potential such as, for example, a screened Coulomb potential (electrons) or a lateral Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction potentials (adatoms).[8] The rapid fall off of the parameter region suggested by the inequalities

$$\begin{aligned}&\varepsilon(1/\nu)\gg\varepsilon(2/\nu),\\ &T\gg\varepsilon(2/\nu)\end{aligned}\tag{7}$$

is of greatest importance. These inequalities imply that one can neglect the next-nearest-electron interactions, and that (as is suggested by our previous discussion) in region (7) only the devil's staircase steps corresponding to the Hubbard structures with $p=1$ can survive at rather large values of the parameter P/T. Thus we will consider just the parameter region (7), taking into account only the interaction between the nearest-neighboring particles. This simplification (NN approximation for short) allows us to construct a self-consistent theory which preserves the most important thermodynamic properties of the 1D system under consideration. Qualitatively the results are insensitive to the concrete form of the repulsion pair potential as is the GWL itself.

III. THERMODYNAMIC POTENTIAL

In the NN approximation the 1D electron system may be considered as an ensemble of independent, noninteracting nearest-neighbor-electron pairs (dimers) which are characterized by interelectron distances $l=1,2,\ldots$ and energies $\varepsilon(l)$. (We will call these l pairs, for short.) The total energy of the system is the sum over all l-pair energies:

$$E=E\{n_l\}=\sum_{l=1}^{\infty}n_l\varepsilon(l),\tag{8}$$

Here $\{n_l\}$ symbolizes a given set of l-pair numbers $n_1,n_2,\ldots$. It must be emphasized that the l-pair ensemble cannot be considered to be an ideal gas of ''particles'' with internal ''quantum numbers'' l and internal energies $\varepsilon(l)$. The reason is that there are *strong space correlations* between l-pair positions. That is transferring a given l pair (typical $l \sim \nu^{-1}$) by a distance $\Delta L \gg \nu^{-1}$ in any direction inevitably results in the order of $\nu \Delta L$ shifts in the opposite direction of other l pairs (by distances $\sim \nu^{-1}$) . It is the combination of these correlations with the additive structure of the energy $E\{n_l\}$ that causes a rather peculiar thermodynamic behavior in the NN approximation. On one hand, the distribution of l pairs over l turns out to be Boltzmann-like (see below), but on the other, the dependencies $\nu(P)$ and $\nu(\mu)$ have nothing in common with those for ideal gases.

To construct the thermodynamics of the system under consideration one must take into account the condition imposed on l-pair numbers to a given system length L, namely,

$$L = \sum_{l=1}^{\infty} l n_l . \tag{9}$$

The simplest way to proceed is to change thermodynamic variables from N, L, and T to N, P, and T, by introducing the isothermal-isobaric partition function

$$Z = Z(N,P,T) = \sum_{\{n_l\}} \exp\left(- \frac{E\{n_l\} + PL\{n_l\}}{T} \right) W\{n_l\}. \tag{10}$$

Here the summation is taken over all possible $\{n_l\}$'s satisfying the condition

$$N = \sum_{l=1}^{\infty} n_l ,$$

where L is the total length given by (9), and

$$W\{n_l\} = N! \bigg/ \prod_{l=1}^{\infty} n_l ! \tag{11}$$

is the statistical weight of the states with a given set of l-pair numbers. Introducing the effective energy

$$\tilde{\varepsilon}(l,P) = \varepsilon(l) + Pl \tag{12}$$

and using formula (11), we can transform expression (10) to the form

$$Z(N,P,T) = \left[\sum_{l=1}^{\infty} \exp\left(- \frac{\tilde{\varepsilon}(l,P)}{T} \right) \right]^N , \tag{13}$$

from which we find that the thermodynamic potential (Gibbs free energy) $\Phi = N\mu(P,T)$ is given by

$$\Phi(N,P,T) = -TN \ln\left[\sum_{l=1}^{\infty} \exp\left(- \frac{\tilde{\varepsilon}(l,P)}{T} \right) \right]. \tag{14}$$

As is to be expected, the thermodynamic potential (14) has quite a different structure from that for ideal gases. It is this which leads to unconventional thermodynamic behavior.

IV. EQUATION OF STATE

First let us find the dependence of the electron density ν on P and T. Using the well-known thermodynamic relation $L = [\partial \Phi(N,P,T)]/\partial P$, from (14) we obtain

$$\nu = \frac{N}{L} = \frac{\sum_{l=1}^{\infty} \exp\left(- \frac{\tilde{\varepsilon}(l,P)}{T} \right)}{\sum_{l=1}^{\infty} l \exp\left(- \frac{\tilde{\varepsilon}(l,P)}{T} \right)}. \tag{15}$$

A similar expression for $\mu(P,T)$ has the form

$$\mu = -T \ln\left(\sum_{l=1}^{\infty} \exp[-\tilde{\varepsilon}(l,P)/T] \right), \tag{16}$$

as follows from (14). Equations of state (15) and (16) establish the links between ν, μ, P, and T. The most convenient choice for the two independent thermodynamic variables is determined by the concrete physical situation under consideration.

A full description of the thermodynamics of the 1D lattice system requires us to amplify the equations of state by determining the average number of l pairs, N_l. According to (13) this quantity is determined by the formula

$$N_l = N(l,P,T) = \frac{\partial \Phi(N,P,T)}{\partial \varepsilon(l)} .$$

Substituting relation (14) into this expression, we find that the distribution of l pairs is of a Boltzmann type; that is,

$$N_l = \frac{N \exp[-\tilde{\varepsilon}(l,P)/T]}{\sum_{k=1}^{\infty} \exp[-\tilde{\varepsilon}(k,P)/T]}. \tag{17}$$

According to (17) the distribution of l pairs is determined by the behavior of the function $\tilde{\varepsilon}(l,P)$, which has an integer-valued argument l. At $P > P_C \equiv \varepsilon(1) - \varepsilon(2)$ this function increases monotonically, while at $P < P_C$ it has a minimum. Due to the properties of the pair potential $\varepsilon(x)$ (see Sec. II) there is only one minimum at integer $l_0 = l_0(P) > 1$, which is located at a distance < 1 from the (single) root of the equation,

$$\frac{d\varepsilon(x)}{dx} = -P. \tag{18}$$

If $P > P_C$, the most probable l-pair length is $l = 1$; while in the opposite case, $P < P_C$, the length is $l_0(P)$.

To analyze (15), (16), and (17) it is convenient to introduce independent variables $\gamma = P/T$ and P, and thus to represent the ratio $\tilde{\varepsilon}(l,P)/T$ in the form $\tilde{\varepsilon}(l,P)/T = \gamma F(l,P)$, by introducing the dimensionless function

$$F(l,P) = \varepsilon(l)/P + l.$$

We now study the limit $\gamma \ll 1$. In this case the main contribution to the sums over l in (15) comes from large values, $l \sim 1/\gamma$, and hence we can replace $F(l,P)$ by simply l. In the $\gamma \to 0$ limit this leads to the classical ideal gas equation of state

$$PL = NT.$$

The result is quite reasonable because in the limit as γ approaches zero the electron-electron interactions is entirely

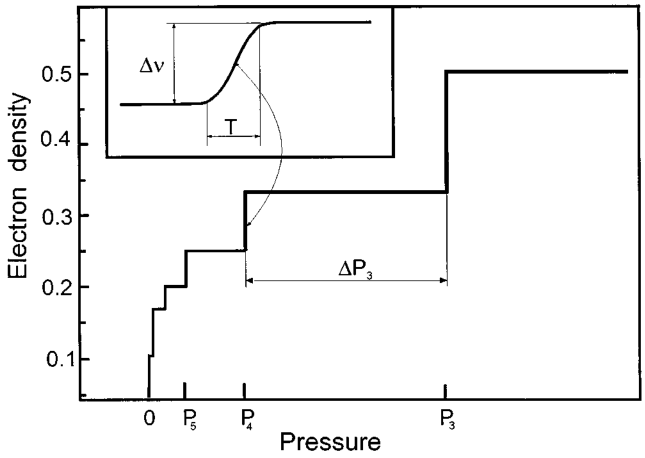

FIG. 1. The dependence of electron density ν against pressure P obtained using (15), $\Delta\nu = 1/(q-1) - 1/q$.

neglected, and the electrons themselves can be considered independent particles. Expanding the sums of (15) in powers of the parameter γ, we obtain the analog of the well-known thermodynamic virial expansion. The first term of it gives the van der Waals–like equation of state,

$$P(L-B(T)N)=TN, \tag{19}$$

where the quantity

$$B(T)=\sum_{l=1}^{\infty}\left[1-\exp\left(-\frac{\varepsilon(l)}{T}\right)\right]>0$$

(essentially, the second virial coefficient) is the effective decrease of the system volume due to the electron-electron repulsion.

In the opposite limiting case, $\gamma \gg 1$, the thermodynamic behavior of the system depends essentially on whether the function $\tilde{\varepsilon}(l,P)$ has a minimum or not (see above). In the first case $(P<P_C)$ only one term, that with $l=l_0(P)$, generally makes the major contribution to sums (15) and (16). More exactly, it takes place for all pressures P which are not too close to the degeneration points P_q determined by the expression

$$P_{q+1}=\varepsilon(q)-\varepsilon(q+1), \quad q=1,2,\ldots . \tag{20}$$

At these values of P the contributions of *two* terms, with $l=[x_0(P_q)]=q$ and $l=[x_0(P_q)+1]=q+1$, coincide $[x_0(P)$ is the root of Eq. (18)]. The set of points $\{P_q\}$ divide the P axis into a set of intervals $\{P_{q+1},P_q\}$, which are (in the framework of the NN approximation) nothing but the devil's staircase intervals corresponding to $p=1$ [see (6)]. The width of the qth interval is

$$\Delta P_q \equiv P_q - P_{q+1} = \varepsilon(q-1) - 2\varepsilon(q) + \varepsilon(q+1).$$

Within these intervals ν is independent of P, with an exponential accuracy by the parameter γ, and equals

$$\nu=\text{const}=1/q. \tag{21}$$

Hence the 1D system has a periodic structure with a period q. This result confirms the suggestion made in Sec. II. Thus, just as in the parameter region $\gamma \gg 1$, for $P<P_C$ the ground-state devil's staircase manifests itself to its full extent (see Fig. 1).

Within the intervals $\{P_{q+1},P_q\}$ the translational symmetry is broken due to thermal creation of l pairs with $l=q\pm 1$. According to (17) the total number of $q\pm 1$ pairs has the exponential dependence

$$\begin{aligned} N_{q-1}&=N\exp\left(\frac{P-P_q}{T}\right), \\ N_{q+1}&=N\exp\left(\frac{P_{q+1}-P}{T}\right). \end{aligned} \tag{22}$$

As follows from (15) and (17), the presence of $q\pm 1$ pairs changes the density ν by a small value,

$$\delta\nu_q=\frac{1}{q^2 N}(N_{q-1}-N_{q+1}). \tag{23}$$

The physical meaning of the last expression is clear. It shows that each $q+1$-pair *increases* the length of the system L by *one* spacing and, conversely, a $q-1$ pair *decreases* L by the same value. These excitations which are elementary ''quanta'' of rarefaction ($q+1$ pairs) or compression ($q-1$ pairs) may be considered as particular kinds of discrete solitons.

From (15) and (16) one easily concludes that the dependence of ν on the chemical potential μ has a similar behavior, the widths of devil's staircase intervals $\Delta\mu_q$ being expressed in terms ΔP_q according to (6). At a given μ the deviation of $\delta\nu$ (23) arises due to a change in the particle number N at a fixed length L. This means that creation on annihilation of *single* electrons in the system increases or decreases, respectively, the total number of the above mentioned rarefaction and compression solitons by q. Hence, in the limit $\gamma \ll 1$, these solitons are effectively free charge carriers with the *fractional* charge

$$e^* = \pm e/q, \tag{24}$$

where e is the electron charge and the signs ''+'' and ''−'' correspond to rarefaction and compression solitons, respectively. Formula (24) generalizes Hubbard's result,[1] which holds for the special case $q=2$. The study of the conductivity of 1D electron systems with LRIR will be the subject of a separate paper.

As one can see from (22) and (23), as P tends to the endpoints P_q, the solitons numbers N_{q-1} and N_{q+1} rapidly increase by an order of magnitude compared with N. This means that the electron density drastically changes between the asymptotic values $\nu=1/(q-1)$ and $\nu=1/q$ upon changing the pressure P by a value $\sim T$. For a description of this

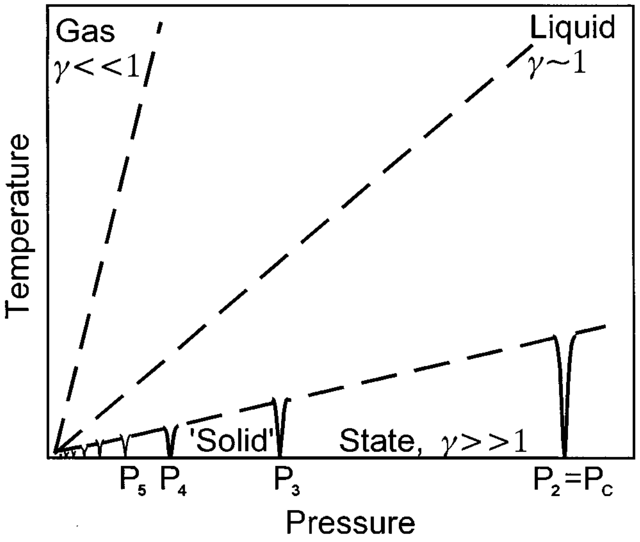

FIG. 2. The schematic distribution of gas $(\gamma \ll 1)$, liquid $(\gamma \sim 1)$ and "solid" $(\gamma \gg 1)$ states in the (P,T) plane. V-shaped curves are conditional boundaries, $\delta\nu_q(P,T)=1/q$ and $\delta\nu_{q+1}(P,T)=1/q$, between the electron liquid and electron "crystal."

intermediate regions one should take into account the two terms with $l=q-1$ and $l=q$ in the sums over l of (15). This leads to the simple formula

$$1/\nu = q - \frac{1}{1+\exp\left(\dfrac{P_q-P}{T}\right)}, \tag{25}$$

which holds over the whole region $|P-P_q| \ll P_q$. When $P_q - P \gg T$ then it reduces to $q-(N_{q-1}/N)$, where N_{q-1} is given by (22). Similarly, in the limit $P-P_q \gg T$, expression (25) turns into $q-1+(N_q/N)$. Within the intermediate domain, $|P_q-P| \sim T$, the system is a highly disordered liquid, since the numbers N_q and N_{q-1} are of the same order of magnitude.

In the case $P > P_C$ and $\gamma \gg 1$ an "electron crystal" with period $q=1$ is formed. The number of translational symmetry-breaking elementary excitations is

$$N_1 = N \exp\left(\frac{P_C - P}{T}\right).$$

These solitonlike excitations are simply electron vacancies with charge

$$e^* = e.$$

Since P_C is also the endpoint P_2 [see (20)], the intermediate region between crystals with $q=1$ and 2 is described by expression (25) at $q=1$.

Thus a change in the pressure P at a given parameter value of $\gamma \gg 1$ produces a succession of electron crystals with periods $q=1,2,\ldots$. The period q increases as P decreases. It should be noted that at fixed $\gamma \gg 1$ these crystals exist even as P tends to zero. In the low-pressure limit the typical value of q is determined by the equation $d\varepsilon(x)/dx|_{x=q} = -P$. The thermodynamic behavior of the system as a whole is schematically represented in Fig. 2.

V. CONCLUSIONS

We have shown that the NN approximation leads to a rich thermodynamics. In spite of the seeming simplicity of the model, it contains the most essential features of discrete 1D systems with LRIR, i.e., strong long-range electron-electron correlations established within the framework of the systems discreteness. It is this that results in the continuous transformation of the 1D electron ensemble from a "gas" state to a "solid," the latter of which is characterized by a set of electron crystals. The parameter $\gamma = P/T$, which controls this transformation, plays the role of a correlation measure. As the temperature decreases, restriction (7) is violated and more and more of the fine structure of the GWL ground state necessarily reveals itself. These tiny steps should appear in the intermediate regions $|P-P_q| \sim T$ located between the large steps of the NN-approximation stair structure. We intend to study this low-temperature problem, which is beyond the scope of the NN approximation, using a general thermodynamic approach. In this context we will study space correlations in 1D LRIR systems (with $t=0$). We hope that the development of our theory will allow us to calculate a number of physical characteristics of the system under consideration, in particular the optical-absorption spectra at finite temperatures (a problem first considered by Hubbard[1]).

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APPENDIX

To derive (6) it is appropriate to consider the changes of the GWL energy $E(N,L)$ $(\nu = N/L = p/q)$ when N and L are decreased or increased by altering the number of electrons per cell, p, or the GWL period, q, respectively. Taking into account the definitions of the devil's staircase endpoints $P_i(\nu)$ and $P_d(\nu)$, we have

$$E(N,L-q) = E(N,L) + qP_d,$$

$$E(N,L+q) = E(N,L) - qP_i. \tag{A1}$$

On the other hand, the energies $E(N,L\pm q)$ can be expressed in terms of the $\nu(\mu)$ devil's staircase endpoints $\mu_i(\nu)$, and $\mu_d(\nu)$ by

$$E(N,L-q) = E(N-p,L-q) + p\mu_i,$$

$$E(N,L+q) = E(N+p,L+q) - p\mu_d. \tag{A2}$$

Increasing N by p and simultaneously increasing L by q is equivalent to inserting one elementary cell into the GWL.

Correspondingly, one can decrease by p and q, which is equivalent to removing one elementary cell. Hence

$$E(N\pm p,L\pm q)=E(N,L)\pm u(\nu), \tag{A3}$$

where $u(\nu)$ is the energy per GWL cell. Combining (A1) and (A2) with (A3), it is easy to show that

$$(\mu_i-\mu_d)p=(P_d-P_i)q.$$

This equality is the same as formula (6) because $\Delta\mu=\mu_i-\mu_d$ and $\Delta P=P_d-P_i$.

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