Ultrametricity, Incommensurability, and Field-Induced Transitions

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The charge- or spin-density-wave formation in one-dimensional electron systems under an incommensurate potential is shown to be characterized by infinitely many order parameters and infinitely many mean-field solutions with almost degenerate energies. The complexity of the solutions is described by ultrametricity. The observed field-induced spin-density-wave transition of 2D systems is discussed in this context.

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The charge- and spin-density-wave states (CDW or SDW) are a manifestation of Fermi surface instabilities. The so-called Peierls theorem ensures that an electron system is always unstable toward density-wave (DW) formation, provided that the Fermi surface satisfies an appropriate nesting condition. This condition is easily satisfied for low-dimensional systems, of which we know a wide variety of examples from inorganic to organic materials. The CDW or SDW states ($2k_F$ Peierls state) in one-dimensional systems are unambiguously described by an order parameter (OP) Δ_Q corresponding to the nesting vector $Q=2k_F$, which is uniquely determined by the Fermi wave number k_F controlled by the electron filling. Whereas a number of OP's may appear in this case, the wave numbers of OP's are limited to the harmonics of $2k_F$. We already understand this situation completely [1].

Here we try to establish the following novel aspects concerning DW formation in one-dimensional electron systems: (1) If we introduce a third length scale δ^{-1}, other than the lattice constant and Q^{-1}, through an external or internal incommensurate periodic potential whose wave number is δ and strength is v, then the stable DW is best described in terms of multiple OP's: OP's of the type $\Delta_N \propto \sum_k \langle C_k^\dagger C_{k+2k_F+N\delta}\rangle$ (N an integer) simultaneously exist to gain the condensation energy. The number of OP's depends on commensurability. In the incommensurate limit it is infinite. This many-OP (MOP) state is more stable than the usual $2k_F$ Peierls state. The realized DW is characterized by a set of OP's, $\{\Delta_N\}$. Under a certain condition, the primary OP, whose amplitude is largest among them, may even differ from Δ_0 with $Q=2k_F$. (2) There exist an abundant or practically infinite number of MOP states as mean-field solutions, each of which satisfies a local minimum condition under a given set of the parameters of the problem. Their OP sequence and magnitudes differ subtly from each other. Some of these states have energies very close to the absolute minimum. (3) The configuration space of the free energy minima is best expressed as having ultrametric topology, reminiscent of those in the so-called complex systems such as spin glasses [2].

We start with the following "generic" mean-field Hamiltonian which describes a one-dimensional system with a tendency toward DW formation under an external periodic potential [3]: $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1$; $\mathcal{H}_0=-\sum_k \cos k\, C_k^\dagger C_k - v\sum_k (C_k^\dagger C_{k+\delta}+\text{H.c.})$, $\mathcal{H}_1=\sum_N \sum_k \Delta_N C_{k+2k_F+N\delta}^\dagger C_k$, with the self-consistent equations $\Delta_N=-U\sum_k \langle C_k^\dagger \times C_{k+2k_F+N\delta}\rangle$, where U is the electron-phonon coupling constant for the CDW and the energy unit is normalized by half the bandwidth. We allow the OP's Δ_N ($N=0, \pm 1, \pm 2, \ldots$).

Let us begin with the simplest example: We consider the quarter-filling case where $2k_F=\pi/2$ and the wave number of the potential $\delta=\pi/4$ (the lattice constant $a=1$). As shown in Fig. 1, the OP's to be taken into account are Δ_{-1}, Δ_0, Δ_1, and Δ_2 in this case because the wave numbers $k+N\delta$ ($N=0$–7) in the first Brillouin zone are coupled to each other through the above Hamiltonian. The matrix elements connecting to these wave numbers give rise to the OP's which have a chance to grow. Simple diagonalization of the 8×8 Hamiltonian matrix readily yields a self-consistent solution with *all* Δ_N *nonvanishing*. For example, when $v=0.02$ ($v=0.04$), $\Delta_{-1}=-0.029$ (-0.055), $\Delta_0=0.035$ (0.037), $\Delta_1=0.006$ (0.014), and $\Delta_2=-0.004$ (0.001) for $U=1.0$. The band splits into 8 bands. We must emphasize that the usual Peierls state with Δ_0 ($Q=2k_F$) and Δ_2 ($Q=4k_F$) *never satisfies* the self-consistent equation in the present $v\neq 0$

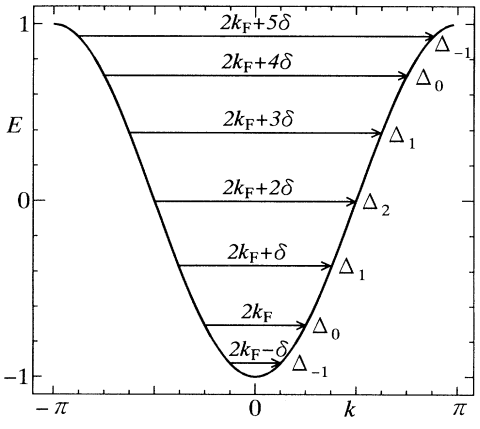

FIG. 1. Relevant wave numbers and associated matrix elements for all possible OP's in the first Brillouin zone for $\delta=\pi/4$.

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situation. Many OP's are inevitable. In order to fulfill it we are bound to take other OP's into account. This is always true irrespective of the filling, δ, v, and U. The above example clearly illustrates that the inevitable infinite chain of coupling among k points occurs when δ^{-1} is incommensurate with the lattice constant. This ultimately leads to the MOP state. The above also shows that the largest OP could be other than Δ_0, in contrast with what is expected by the Peierls theorem, that the filling uniquely determines the nesting vector $Q=2k_F$.

There is a MOP state which continuously grows from the usual Peierls state as $|v|$ increases from zero. In this MOP state, we can estimate $\Delta_{N\neq 0}$ perturbationally: The Nth OP contributes to opening up the gap at the Fermi level in the Nth-order process in $|v|$. By this estimate, we see that $|\Delta_N/\Delta_0|=c_N v^{|N|}$ and the proportionality constant c_N becomes large as δ becomes smaller because v is scaled by δ in the present situation. This result implies that as $|v|$ increases or δ decreases, the largest OP will shift successively from Δ_0 to $\Delta_{\pm 1}$, $\Delta_{\pm 2}$, $\Delta_{\pm 3}$, and so on. We have found such a MOP state numerically and confirmed the above statements [4].

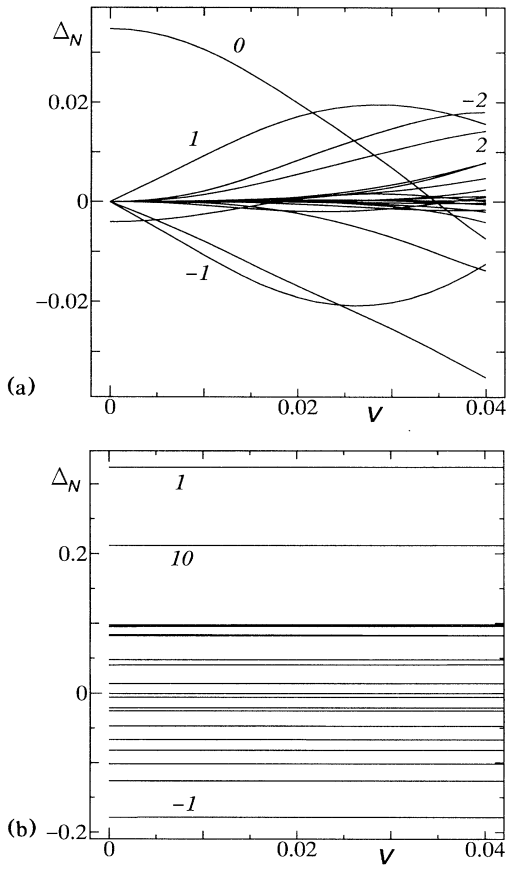

FIG. 2. Example of the mean-field solutions as a function of v ($p=40$ and quarter-filling case). (a) The perturbational solution where various OP's change over smoothly. The largest OP interchanges successively ($U=1.0$). (b) A nonperturbational solution where each OP does not change and persists down to $v=0$ ($U=2.0$).

However, if we approach the incommensurate situation in the above numerical calculation, namely, if p becomes large where $\delta=2\pi r/p$ (the integers p and r are mutually prime, p is called the commensurability index), the problem becomes nontrivial and further new aspects emerge. When $p=40$ and $r=1$, for example, the number of possible independent OP's is 20, and we must solve the 40×40 Hamiltonian matrix self-consistently. To find a local minimum solution we employ a numerical iteration method starting with an initial set of the OP's. Although there exists a solution corresponding to the above perturbation theory, we have found an abundant number of solutions with comparable energies. One example of our calculations is shown in Fig. 2. Note that as $|v|$ decreases the perturbational solution (a) smoothly reduces to the usual $2k_F$ Peierls state while the solution (b) does not and persists down to $v=0$. Each solution in which many OP's coexist is characterized by a sequence of the twenty OP's, $\{\Delta_N\}$.

The solutions obtained are conveniently classified into a hierarchical tree-like structure in which not only the primary OP's differ from each other, which forms the first "generation," but the second or third generation OP's differ as shown in Fig. 3. Namely, two states $\{\Delta_1,\Delta_2,\Delta_3,\ldots\}$ and $\{\Delta_1,\Delta_2,\Delta_4,\ldots\}$ differ only from the third OP and branch at the "second generation," leading to an infinitely iterating tree structure. In principle, there can exist $p!$ different states although in practice the computer algorithms of the simple iteration method employed prevent obtaining all possible solutions. (According to our experience larger U or $|v|$ make many local minima easier to be found.) It is an extremely difficult task to find the absolute minimum solution among such an abundance of metastable ones because their energies are almost degenerate.

Physically, we can express this complexity of our prob-

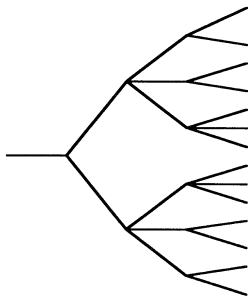

FIG. 3. Schematic structure of mean-field-solution space where the solutions are classified according to the ordering of many OP's. The end points on the right-hand side correspond to particular solutions. Branching of various solutions is characterized by the sequence of many OP's, $\{\Delta_N\}$, yielding a hierarchical subphase structure in real materials.

lem as follows: In order to maximally gain the DW condensation energy, each OP which has a chance to grow tries to open up the maximal energy gap right at the Fermi level directly or indirectly. However, some of the OP's are mutually depressing and some are enhancing collectively. This hidden frustration originates from the incommensurability of the potential where every site is subjected to different potential values and there is no identical site throughout the system. This situation is quite similar to a situation in spin glasses or other combinatorial optimization problems [2]. In fact, the resulting DW pattern in real space looks quite "glass" like. It is evident from the hierarchical tree structure that our configuration space is ultrametric [2]. The distance $d(\alpha,\beta)$ between two solutions α and β defined to be proportional to the "generation" of their closest common ancestor satisfies the ultrametric inequality: $d(\alpha,\beta) \leq \mathrm{Max}\{d(\alpha,\gamma),d(\beta,\gamma)\}$.

To further qualify our assertion on this point let us consider statistics of the configuration space of our solutions. We first define a distance or overlap between two solutions α and β as $q^{\alpha,\beta}=[\sum_{i=1}^{p}(\Delta_i^{\alpha}-\Delta_i^{\beta})^2]^{1/2}$, where p is the commensurability index introduced earlier. By taking a triplet (α,β,γ) among the obtained solutions the distances are normalized as $q_i^R \equiv 3q_i/(q_1+q_2+q_3)$ $(i=1,2,3,)$, where $q_i=q^{\alpha,\beta}$, $q^{\beta,\gamma}$, or $q^{\gamma,\alpha}$. Then, we check the ultrametricity of our configuration space. As shown in Fig. 4 the statistics satisfy well the ultrametric inequality, implying that the free energy landscape is ragged and the ordered DW phase is marginally stable [2]. This feature of our problem leads to specific experimental consequences: In particular, the dynamics toward equilibrium differ drastically from the ordinary ordered phases such as the conventional Peierls state, which are characterized by slow relaxational dynamics described by the stretched exponential type or the power-law type, depending on the detailed structure of our ultrametric space. We also expect characteristic hysteresis phenomena, which have already been emerging experimentally as explained below. A slight change of the parameter of the problem by magnetic field or by pressure leads to a drastic change of the stable DW state, therefore resulting in the fine subphase structure in the phase diagram.

Let us consider several applications of our assertions to real low-dimensional materials. The best example is the so-called field-induced SDW (FISDW) phenomena observed in Bechgaard salts, $(TMTSF)_2X$ $(X=ClO_4,PF_6)$ [5], a problem of SDW formation in a strongly anisotropic two-dimensional electron system under a perpendicular field. It is known [6] that such a system can be cast into a one-dimensional form and described by starting with the Harper equation which is nothing but $\mathcal{H}_0$. In this case $\delta=(2\pi/a)\phi/\phi_0=(2\pi/a)eabH/2\pi c$, where ϕ is the flux per unit plaquette, b the transverse lattice constant, ϕ_0 the flux quantum, and v the transverse hopping integral [6]. [Note in passing that v is scaled by $v_F\delta(H)$.

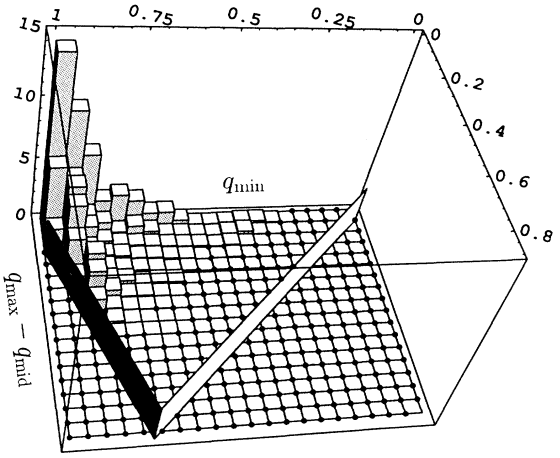

FIG. 4. Histogram of the distribution of the distances for 27 solutions for $v=0.35$, $p=60$, $U=3.0$, and next neighbor hopping 0.2 for SDW (total triplets are 2925), where $q_{\min}$, q_{mid}, and $q_{\max}$ are three distances for a triplet. The ultrametric inequality is satisfied when the distribution is along the $q_{\min}$ axis, while the triangle represents the usual triangular inequality.

This point is important when applying our theory to the experiments.] Thus, interestingly enough, we can control continuously the incommensurability through magnetic field ($\delta\sim$ the order of 10^{-3} for $H\sim 30$ T). Experiments [7] reveal a recursive H vs T phase diagram: Finer and finer subphases emerge as T decreases. This is quite understandable if we look at the hierarchical tree shown in Fig. 3. The decreasing T sweep corresponds to moving on this tree from left to right [2]. Therefore we can understand the unexplained facts that the FISDW phenomena are cooling dependent and that marked hysteresis is found. Upon decreasing T the cooling rate determines decisively the "branching" in the hierarchical tree and therefore the realized low temperature phase would differ widely depending upon the cooling history. This explains the different results of quantized Hall conductance measurements, which include abrupt sign changes of the Hall voltage [5]. As for the finer subphases or recursive phase diagram, it is quite expected now because as H varies, or the incommensurability $\delta(H)$ continuously changes, different ground states are realized for every H. None of them coincide. The H vs T phase diagram is divided into infinitely many subphases, resulting in a recursive one if the cooling rate is infinitely slow [8]. A physical reason that the finer structure is observed in spite of "weak" field or extremely large incommensurability lies in the fact that the effective unit cell of the FISDW is enlarged because of the incommensurate Q vector.

It is known [9] that by starting from the strong field limit and taking the lattice potential as a small perturbation, the same Harper equation is recovered as in the weak field. Thus our assertions are applicable to this case by appropriately reinterpreting the parameters (e.g., δ

$\propto H^{-1}$). This enables us to discuss systems in the quantum limit in contrast with the first example of a tight-binding Hamiltonian where H is weak compared to the lattice potential. Graphite, which is semimetallic, is known to exhibit a CDW transition under perpendicular fields [10] around $H=30$–40 T, at which point the quasi quantum limit is achieved at low temperatures. This quasi-two-dimensional material is a good candidate to test our assertions, in particular, to investigate the possibility of the recursive phase diagram associated with the MOP state of the CDW phase. It seems that some experiments [10] are revealing a complex phase diagram.

One of the important aspects of our assertions is that the $2k_F$ DW system is always unstable against the MOP state when H is applied, where the primary OP may differ from Δ_0 with $Q=2k_F$ and, moreover, many subsidiary OP's are spontaneously induced, resulting in a novel DW state. In this connection we point out that the high field phase in another low-dimensional charge transfer organic conductor, $(BEDT\text{-}TTF)_2KHg(SCN)_4$ [11], in which the $2k_F$ SDW appears at $T=10$ K under no applied field, might be changed over to a MOP state. By applying a magnetic field we can drive the $2k_F$ SDW to a MOP state.

The recent advances of material design on artificial two-dimensional systems have great promise not only to check our assertions, but also to find a new phenomenon associated with the frustration due to competing length scales. In fact, a novel magnetoresistance oscillation in a periodically modulated two-dimensional system in an AlGaAs/GaAs heterojunction is emerging [12].

The MOP state itself has been already pointed out to exist by Machida and co-workers [13] and independently by Lebed' [14] within a specialized model suitable for the FISDW in quasi-two-dimensional organic conductors. Our newly gained viewpoints here are summarized as follows: (a) In order to demonstrate the generality of the concept of the MOP state, we have succeeded in reducing the previous specific model to a "minimal and generic" model system to produce the MOP state both for CDW and SDW. This allows us to discuss the applicability of the MOP state to various research fields other than the FISDW in a broader perspective. In other words, we have revealed what conditions are needed for the MOP state to occur. (b) We have shown explicitly for the first time the "complexity" of our problem associated with the MOP state. We have emphasized that this aspect is the essence of the problem. This recognition might be important to perform and interpret experiments in these fields and shed light on the linkage of the present problem to seemingly remote research fields such as spin glasses, which enables us to gain a new perspective on our problem.

In conclusion, we have pointed out a novel aspect of the CDW and SDW formations under an incommensurate potential. Application of a magnetic field to low-dimensional conductors is an ideal means to test our proposals.

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