Phase separation due to quantum mechanical correlations

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Can phase separation be induced by strong electron correlations? We present a theorem that affirmatively answers this question in the Falicov-Kimball model away from half-filling, for any dimension. In the ground state the itinerant electrons are spatially separated from the classical particles.

The Falicov-Kimball (FK) model [1] can be viewed as a modification of the Hubbard (H) model [2] in which one species of electrons (say spin down) has infinite mass. As such, its relation to the latter is similar to the relation of the Ising model to the quantum Heisenberg model [3]. Alternatively, it can be viewed as a model of itinerant electrons and immobile ions. It possesses long range order at low temperature in two or more dimensions at half filling, and this checkerboard state (and higher-period generalizations [4–7]) remain to-date as the only examples of crystallization into a perfectly ordered structure whose periodicity is not that of the underlying lattice.

In this paper, we report a theorem on the existence of phase separation in the ground state of the FK model, away from half-filling and for large repulsion between the particles. 'Phase separation', or 'segregation', means that the system splits into two large domains, one being occupied by the classical particles, and the other by the quantum particles. In the language of the H model, this would mean segregation of spin up particles from spin down particles — resulting in a ferromagnetic state. The question of whether strong interactions can drive quantum (electronic) systems to phase separate was posed over ten years ago for the FK model [8] and the H model [9]. This work is a rigorous proof of this long-standing conjecture for the FK model. Further discussion of the relation between the FK and the H model is given later.

Phase separation is not new in classical lattice models. It is present in the Ising model, and in many other classical models. It also occurs in the FK model at half-filling for some densities, as was proved in [6]. In these examples it is mainly a *local* phenomenon — interactions (or 'effective interactions' in the case of FK) tend to dislike boundaries, and the state with minimum boundary is phase separated.

Away from half-filling, the electrons of the FK model are in delocalized wave functions, and their energy cannot be written as a sum of local terms. While we prove that the ground state energy is roughly proportional to the boundary between occupied and empty sites, the mechanism is *nonlocal* and genuinely quantum mechanical.

The FK Hamiltonian [1] is

$$H = -\sum_{\mathbf{x},\mathbf{y}\in\Omega} t(\mathbf{x}-\mathbf{y}) c^\dagger_{\mathbf{x}} c_{\mathbf{y}} + U \sum_{\mathbf{x}\in\Omega} c^\dagger_{\mathbf{x}} c_{\mathbf{x}} w_{\mathbf{x}}. \tag{1}$$

Here, $t(\mathbf{x}-\mathbf{y})$ is the hopping coefficient between sites $\mathbf{x}$ and $\mathbf{y}$; it is translation invariant, but may depend on the direction (this allows consideration of general Bravais lattices). $c^\dagger_{\mathbf{x}}$ and $c_{\mathbf{x}}$ are creation and annihilation operators for a spinless electron at site $\mathbf{x}$, and $w_{\mathbf{x}} = 1$ or 0 is a classical variable that denotes the presence or the absence of an ion at $\mathbf{x}$. (Spin degrees of freedom have trivial behavior and are left aside here.) $\Omega \subset \mathbb{Z}^d$ is a finite d-dimensional lattice, and U is the on-site repulsion between the two species of particles. For any given configuration $w = \{w_{\mathbf{x}}\}$ of classical particles, the ground state for $N_{\rm e}$ electrons is determined by diagonalizing a one-body operator given by the above Hamiltonian, and filling in the lowest $N_{\rm e}$ states. The main question is to find which configuration w, with a given number of classical particles $N_{\rm c} = \sum_{\mathbf{x}} w_{\mathbf{x}}$, minimizes the energy of the electrons.

Our theorem states upper and lower bounds for the energy of $N_{\rm e}$ electrons, for a given configuration w of the classical particles. For orientation, let us consider first a configuration where the sites devoid of classical particles form a large, 'compact' region. The expected energy of the electrons is a bulk term that scales like the volume of this region, and a correction that scales like its boundary. Our main result is a proof of this conjecture for *all* configurations, not only 'nice' ones.

We need some notation. Let $\Lambda = \{\mathbf{x} \in \Omega : w_{\mathbf{x}} = 0\}$ denote the set of empty sites for the configuration w, and $\partial\Lambda$ its boundary, $\partial\Lambda = \{\mathbf{x} \in \Lambda, \mathrm{dist}(\mathbf{x}, \Omega \setminus \Lambda) = 1\}$. Their respective number of sites are $|\Lambda|$ and $|\partial\Lambda|$. We write $E(N_{\rm e}, w)$ for the ground state energy of $N_{\rm e}$ electrons in the configuration w. An important quantity is $n = n_{\rm e}/(1-n_{\rm c})$, where $n_{\rm e} = N_{\rm e}/|\Omega|$ and $n_{\rm c} = \sum_{\mathbf{x}} w_{\mathbf{x}}/|\Omega|$ are the densities for quantum and classical particles respectively. It represents the electronic density that would exist inside Λ, if all electrons live inside the domain devoid of classical particles. Let $e(n)$ be the usual kinetic energy per site for noninteracting electrons with density n in the thermodynamic limit (its expression is recalled below, see Eq. (3)).

Theorem. *For all Λ we have upper and lower bounds, $e(n)|\Lambda| + \alpha'(n)|\partial\Lambda| \geq E(N_e, w) \geq e(n)|\Lambda| + \alpha(n,U)|\partial\Lambda|$.*

Here, $\alpha'(n)$ and $\alpha(n,U)$ are explicitly given positive functions. For nearest-neighbor hoppings (i.e., $t(\mathbf{x}) \neq 0$ if $|\mathbf{x}| = 1$, $t(\mathbf{x}) = 0$ otherwise), $\alpha(n,U) = \alpha(n) - \gamma(U)$, where $\alpha(n) = \alpha(1-n)$ is strictly positive for $0 < n < 1$, and $\gamma(U)$ satisfies $\lim_{U\to\infty} U\gamma(U) = 8d^2$.

The theorem clearly leads to segregation because low-energy configurations must have a small boundary, i.e. there is a relatively small number of empty sites that are neighbors of the classical particles. Furthermore, due to the large repulsion, electrons are essentially located in the empty domain. The boundary of the configuration of classical particles that minimizes the energy is smaller than the minimum possible boundary times the ratio between the upper and lower bounds (which is a large constant).

Phase separation in the FK model for large U was conjectured in [8] and is in stark contrast to the situation at $n_c = n_e = \frac{1}{2}$, where long-range order of checkerboard type occurs, as was established in [3]. The theorem is proved in [10], and extends results for $d = 1$ [11] and $d = \infty$ [12] to all dimensions, in particular to the dimensions 2 and 3 that are of great physical relevance. Its proof relies on a result of Li and Yau for the Laplace operator in the continuum [13] (see also [14], Theorem 12.3). We explain this elegant proof below in the case of a lattice, thereby proving the lower bound for $U = \infty$ without the boundary correction. To include this boundary correction requires considerably more effort, and we refer to [10] for further details.

Proof with $\alpha(n) = 0$ and $U = \infty$: The ground-state energy $e(n)$ for noninteracting electrons with density n is found in the usual fashion: (i) define a Fermi energy ε_F via

$$\frac{1}{(2\pi)^d}\int_{\epsilon(\mathbf{k})\leq\varepsilon_F} \mathrm{d}^d k = n, \tag{2}$$

with the band structure $\epsilon(\mathbf{k}) = -\sum_{\mathbf{x}} t(\mathbf{x})\exp(-\mathrm{i}\mathbf{k}\cdot\mathbf{x})$ and (ii) do the integration

$$\frac{1}{(2\pi)^d}\int_{\epsilon(\mathbf{k})\leq\varepsilon_F} \epsilon(\mathbf{k})\,\mathrm{d}^d k = e(n). \tag{3}$$

The electrons are forbidden to lie on any site occupied by the classical particles. Then the eigenfunctions for a given configuration w are found by diagonalizing the projection of the hopping matrix onto Λ. Let $\phi_\beta(\mathbf{y}; w)$ denote the orthonormal eigenvectors of the Hamiltonian indexed by $\beta = 1, ..., |\Omega| - N_c$ for the configuration w with eigenvalues $\epsilon_\beta(w)$. We choose the ordering of the labels such that $\epsilon_1(w) \leq \epsilon_2(w) \leq ... \leq \epsilon_{|\Omega|-N_c}(w)$. We also extend the definition of the eigenvectors to all of $\mathbb{Z}^d$ setting $\phi_\beta(\mathbf{y}; w) = 0$ for all $\mathbf{y} \notin \Omega$. The ground state energy for N_e electrons and N_c classical particles in the configuration w is

$$E(N_e, w) = \sum_{\beta=1}^{N_e} \epsilon_\beta(w), \tag{4}$$

and the ground-state energy $E_{\mathrm{g.s.}}(N_e, N_c)$ is the minimum of Eq. (4) over all configurations that contain N_c classical particles.

Using the definition of the eigenvectors allows us to write the ground state energy as

$$E(N_e, w) = \sum_{\beta=1}^{N_e} \sum_{\mathbf{y},\mathbf{z}\in\mathbb{Z}^d} \phi^*_\beta(\mathbf{y}; w)[-t(\mathbf{y}-\mathbf{z})]\phi_\beta(\mathbf{z}; w). \tag{5}$$

Now, we define the Fourier transform of the eigenfunctions by

$$f_\beta(\mathbf{k}; w) = \sum_{\mathbf{y}\in\mathbb{Z}^d} \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{y}}\phi_\beta(\mathbf{y}; w), \tag{6}$$

for every wave vector $\mathbf{k}$ in the Brillouin zone $[0, 2\pi]^d$. It is well-known that the energy can be expressed as

$$E(N_e, w) = \frac{1}{(2\pi)^d}\int \mathrm{d}^d k\, \epsilon(\mathbf{k})\rho(\mathbf{k}; w); \tag{7}$$

here and in the following the integral is over $[0, 2\pi]^d$ and we introduced the density function

$$\rho(\mathbf{k}; w) = \sum_{\beta=1}^{N_e} |f_\beta(\mathbf{k}; w)|^2. \tag{8}$$

The density function is obviously positive and is bounded above by $|\Lambda|$. Indeed, we can write

$$\rho(\mathbf{k}; w) = \sum_{\beta=1}^{N_e} \sum_{\mathbf{y},\mathbf{z}\in\mathbb{Z}^d} \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{z}}\phi^*_\beta(z; w)\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{y}}\phi_\beta(y; w); \tag{9}$$

this can be rewritten as

$$\rho(\mathbf{k}; w) = \sum_{\mathbf{y},\mathbf{z}\in\Lambda} \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{z}}\rho(\mathbf{z}, \mathbf{y}; w)\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{y}} \tag{10}$$

with $\rho(\mathbf{z}, \mathbf{y}; w) = \sum_{\beta=1}^{N_e} \phi^*_\beta(\mathbf{z}; w)\phi_\beta(\mathbf{y}; w)$. The matrix ρ is a positive semidefinite matrix bounded by 1 (it is the projector onto the lowest N_e eigenvectors). Hence,

$$\rho(\mathbf{k}; w) \leq \sum_{\mathbf{y}\in\Lambda} |\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{y}}|^2 = |\Lambda|. \tag{11}$$

Furthermore, the density function satisfies a sum rule

$$\frac{1}{(2\pi)^d}\int \mathrm{d}^d k\, \rho(\mathbf{k}; w) = \sum_{\mathbf{y}\in\Lambda} \rho(\mathbf{y}, \mathbf{y}; w) = N_e. \tag{12}$$

One gets a lower bound for $E(N_e, w)$ by minimizing the right side of (7) over all functions ρ satisfying $0 \leq \rho \leq |\Lambda|$ and whose integral is N_e. This is the 'bathtub principle', see [14], Theorem 1.14; the minimizer is $\rho(\mathbf{k}) = |\Lambda|$ for $\{\mathbf{k} : \epsilon(\mathbf{k}) \leq \varepsilon_{\rm F}\}$ and $\rho(\mathbf{k}) = 0$ otherwise, with $\varepsilon_{\rm F}$ defined by Eq. (3). This amounts to filling the lowest eigenvalues of the infinite-lattice.

The proof of the upper bound for the energy proceeds by forming an 'average' Hamiltonian by translating and rotating the original configuration w over a large but finite subset of $\mathbb{Z}^d$ (with periodic boundary conditions). Then on a bipartite lattice, one can show by concavity of the sum of the lowest N eigenvalues of a matrix, that the averaged Hamiltonian provides an upper bound to the ground state energy. But the magnitude of the averaged hopping is determined by the size of the boundary, which eventually yields the desired upper bound. Extending the proof above to provide the lower bound is much more complicated and relies on a detailed technical examination of the influence of the boundary sites on the minimal density function for a given configuration of classical particles. It is done in the isotropic case (that is, a hypercubic Bravais lattice with equal hoppings in all directions) in [10]; the extension to the anisotropic case is straightforward, but numerous details are modified, and have been verified.

The results of the theorem have a number of implications for the FK model. It establishes that the segregation principle holds in all dimensions (at $T = 0$ and $U = \infty$) illustrating the fact that the existence of periodic ground states requires a subtle reduction in energy relative to the segregated phase as the interaction strength is made finite. Since the electronic wave functions will be exponentially localized within Λ, the results shown here can be extended to the case of finite interaction strength, as long as U is large enough. At positive temperature it is so far impossible to claim rigorous results, except the following weaker one: the electronic free energy (for a *fixed* configuration of classical particles) can be shown to be equal to the bulk free energy plus a correction term that is proportional to the size of the boundary. We expect that the coexistence of two phases occurs at finite temperature for $d \geq 2$ as happens in the Ising model.

It may be instructive to consider the $U = \infty$ results for the FK model as a guide for possible behavior in the H model. To do this, we must first find a way to interpolate between the two models. The simplest way is to consider an asymmetric-hopping H model where the hopping for the spin-up and the spin-down particles is different. Then the H model results when $t_\uparrow = t_\downarrow$ and the FK model when $t_\downarrow = 0$. Our rigorous results only hold for $t_\downarrow = 0$. When $t_\downarrow$ is increased the classical particles should still be packed, due to the pressure of the electrons. When $t_\downarrow$ keeps increasing however, the classical particles should be in a phase with density strictly less than 1. The central issue is whether the reduction of the down-spin kinetic energy can be made large enough, so that the phase separation disappears at a critical value of $t_\downarrow$. If this occurs for all electron densities, there is no (saturated) ferromagnetism in the $U = \infty$ H model on the given lattice; however, one has a saturated ferromagnetic ground state if the phase separation survives (note the $SU(2)$-imposed degeneracy of the ferromagnetic multiplet will occur precisely at $t_\downarrow = t_\uparrow$). It is well known that ferromagnetism depends strongly on the geometry of the lattice [15,16], so the occurrence of a critical value of $t_\downarrow$ must also depend strongly on the geometry of the lattice. We are unable to make any rigorous statements about ferromagnetism in the H model here — actually, we do not even know how to study the case with nonzero, but small $t_\downarrow$.

A major question is what happens to the chessboard phase when doped away from half filling? Consider the line $n_{\rm e} = n_{\rm c}$; the chessboard phase is present when these densities are equal to $\frac{1}{2}$, and segregation takes place when they differ significantly from $\frac{1}{2}$ (depending on U). It is not clear what to expect for intermediate values. Two possible scenarios are (i) the coexistence between chessboard and segregated phases or (ii) the coexistence between other periodic and segregated phases. Both scenarios could be of physical relevance to stripe physics.

We conclude this letter by a summary of our knowledge of the phase diagram for zero temperature and $d \leq 2$. Recall that the particle-hole symmetry implies that the phase diagram is symmetric under the transformation $(n_{\rm e}, n_{\rm c}) \mapsto (1 - n_{\rm e}, 1 - n_{\rm c})$ [3]. In the sequel we describe the situation for $n_{\rm e} + n_{\rm c} \leq 1$, which is enough.

In two dimensions, the ground states are **periodic**

- when $(n_{\rm e}, n_{\rm c}) = (\frac{1}{2}, \frac{1}{2})$ (they are of the chessboard type). This was proved for all U in [3];
- when $n_{\rm c} = 1 - n_{\rm e}$ (i.e. at half-filling), and $n_{\rm e} = \frac{2}{5}, \frac{1}{3}, \frac{1}{4}, \frac{2}{9}, \frac{1}{5}, \frac{2}{11}, \frac{1}{6}$; also, $n_{\rm e} = \frac{1}{n^2 + (n+1)^2}$ with integers n. This holds for U large enough (depending on $n_{\rm e}$), and follows from [4–6,17,18];
- when $n_{\rm c} = 1 - n_{\rm e}$, and the electronic density $n_{\rm e}$ is a rational number between $\frac{1}{3}$ and $\frac{2}{5}$. U must be larger than a value that depends on the denominator of $n_{\rm e}$ [7].

There is **coexistence** of two periodic phases

- when $n_{\rm c} = 1 - n_{\rm e}$ and $n_{\rm e} \in (\frac{1}{6}, \frac{2}{11}) \cup (\frac{1}{5}, \frac{2}{9}) \cup (\frac{2}{9}, \frac{1}{4})$, for U large [6,17].

And the ground states display **segregation**

- for $\frac{\rm const}{U}(1 - n_{\rm c}) < n_{\rm e} < (1 - \frac{\rm const}{U})(1 - n_{\rm c})$; this is described here, and proved in [10].

These results are illustrated in Fig. 1.

The domain $\frac{\rm const}{U}(1 - n_{\rm c}) > n_{\rm e}$ should also be segregated. The central band that includes the line $n_{\rm e} + n_{\rm c} = 1$ should be the host of numerous periodic phases and various coexistences between periodic phases and empty or

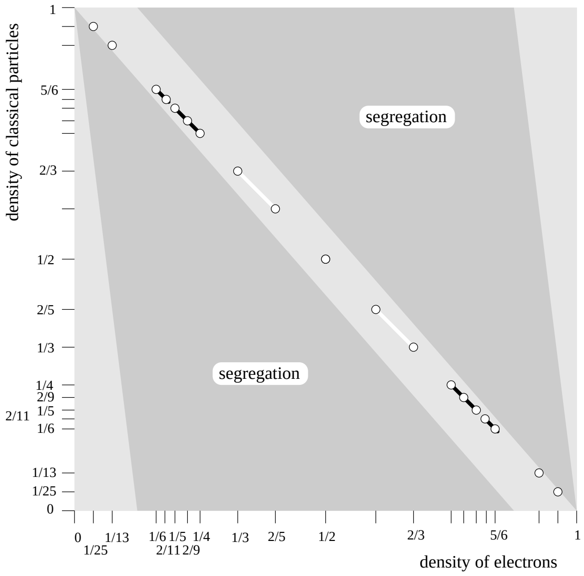

FIG. 1. Schematic phase diagram of the rigorous results for the ground state of the 2D Falicov-Kimball model for large U. The white dots and the white lines represent periodic phases; the black lines are coexistences between different periodic phases; and dark gray regions are segregated. There are no rigorous results for light gray domains.

full phases. This is supported by numerical simulations in 2D [19].

In one dimension, the ground states are **periodic**

- when $(n_{\rm e}, n_{\rm c}) = (\frac{1}{2}, \frac{1}{2})$ [3];
- when $n_{\rm c} = 1 - n_{\rm e}$ (half-filling) and $n_{\rm c} = p/q$ is a rational number (in an irreducible fraction). The periodicity is q for U sufficiently large (depending on q) [11].

For finite U, there is numerical evidence for **coexistence** of two periodic phases and free electrons

- when $n_{\rm c} = 1 - n_{\rm e}$, $p/q < n_{\rm c} < p'/q'$, and the periodic phases with period q and q' are the only stable phases within the above interval [20].

And the ground states display **segregation**

- when $n_{\rm e} \neq 1 - n_{\rm c}$ and U is sufficiently large [11,10].

The canonical phase diagram for small U is even richer but our knowledge of it is very limited.

In conclusion, we have proved that the FK model is phase separated whenever $N_{\rm e} < |\Lambda|$ and $U \to \infty$. This shows how strong correlations can lead to phase separation.

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