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LETTER TO THE EDITOR

A complete devil's staircase in the Falicov–Kimball model

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Abstract. We consider the neutral, one-dimensional Falicov–Kimball model at zero temperature in the limit of a large electron–ion attractive potential, U. By calculating the general n-ion interaction terms to leading order in $1/U$ we argue that the ground state of the model exhibits the behaviour of a complete devil's staircase.

In this letter we study the ground-state phase diagram of the one-dimensional Falicov–Kimball model. This model was proposed to describe metal–insulator transitions [1] and has since been investigated in connection with a variety of problems such as binary alloys [2], ordering in mixed-valence systems [3], and the formation of ionic crystals [4]. It is the latter language we shall use here, considering a system of static positive ions and mobile spinless electrons. The model comprises no electron–electron or ion–ion interactions but an on-site electron–ion attraction, $-U$.

We write the Falicov–Kimball model in the form

$$\begin{aligned}\mathcal{H} = t\sum_j (a_j^\dagger a_{j+1} + a_{j+1}^\dagger a_j) - U\sum_i (s_j a_j^\dagger a_j - 1/2) + (U/2 - \mu_{\mathrm{i}})\sum_j (s_j - 1/2) \\ + (U/2 - \mu_{\mathrm{e}})\sum_j (a_j^\dagger a_j - 1/2)\end{aligned} \tag{1}$$

where $a_i^\dagger$ (a_i) denotes the fermionic creation (destruction) operator for a spinless electron, s_i is equal to 1 (0) if site i is (un)occupied by an ion, t is the hopping integral for electrons, μ_{i} and μ_{e} are the chemical potentials for ions and electrons, respectively, and U is a positive constant corresponding to the ion–electron attractive energy. The choice of a positive U is not restrictive since the transformation $\{U \to -U; \mu_{\mathrm{i}} \to -\mu_{\mathrm{i}}; s_i \to 1 - s_i\}$ maps the Hamiltonian (1) onto the same system with U negative.

The ground state of the system is chosen by minimizing the energy per site over all possible ionic arrangements. The structure of the ground states differs significantly depending on whether U is large or small compared to t. In the first case the electrons are essentially localized near the ions and the latter tend to be as far apart as possible while, for large t/U, the delocalization of electrons favours the formation of clusters of ions [5]. In this letter we consider the case where U is very large compared to other parameters in (1), and treat t/U as a perturbative parameter.

Despite the simplicity of the Falicov–Kimball model the determination of the ground state is far from trivial. Numerical results [6] have suggested that in the neutral system, where the number of electrons and ions are equal, a large number of modulated phases appear as ground states. In 1989 Barma and Subrahmanyam studied the phase diagram of the model by mapping it onto an Ising system [7]. They showed that the phases appearing at the first few stages of a perturbative analysis could be described in terms of a simple branching rule, hence suggesting that the complete phase diagram might display a devil's staircase, an infinite sequence of commensurate (and possibly incommensurate) phases [8]. A different approach to the large-U limit was later introduced by Gruber *et al* [4] who considered the model as a set of ions with interactions mediated by the electrons. They calculated the two-ion interaction to leading order in t/U on the basis of which they argued that the ion spacing is constant in the ground state.

Here we show that a full determination of the ground state requires a calculation of the general m-ion interactions. These are obtained to leading order in t/U using Green's function techniques. Then, using arguments first introduced by Fisher and Szpilka [8], we deduce the existence of a devil's staircase in the neutral Falicov–Kimball model. To our knowledge this is the first quantum model which has been shown to have this behaviour.

The phase diagram for $t = 0$ is shown in figure 1. All the phase boundaries in the figure are multi-degenerate in that any phase obtained by mixing the two neighbouring phases is degenerate on the boundary. Our aim is to study systematically how this multi-degeneracy is lifted as t/U increases from zero.

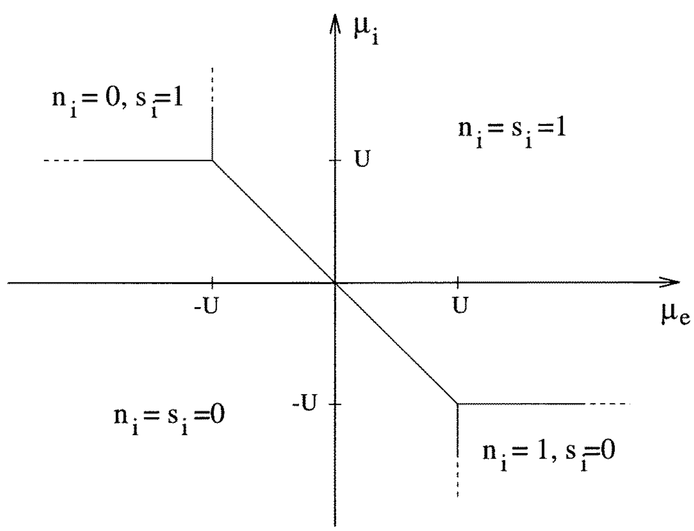

Figure 1. The phase diagram of the Falicov–Kimball model for $t = 0$.

It is convenient to introduce the variables

$$h \equiv (\mu_\mathrm{i} + \mu_\mathrm{e})/2 \qquad \Delta \equiv (\mu_\mathrm{i} - \mu_\mathrm{e})/2. \tag{2}$$

U is assumed to be much larger than any physical parameter in (1) and therefore $\Delta/U \ll 1$. This restriction on Δ has the important consequence of fixing the total number of electrons equal to the total number of ions, and throughout the rest of the paper, we will implicitly consider a neutral system, $\sum_i n_i = \sum_i s_i$, where $n_i = a_i^\dagger a_i$.

When moving along the line $\mu_\mathrm{e} = \mu_\mathrm{i}$ in figure 1 one notices that, for negative values of h, the ground state corresponds to an empty lattice ($n_i = s_i = 0$). On the other hand, for h positive $n_i = s_i = 1$. The point $h = 0$ lies on the multi-degenerate phase boundary where all phases associated with an arbitrary spacing of the ions are degenerate. To distinguish between the different degenerate states it is convenient to introduce the labelling $\langle n_1, n_2, \ldots, n_m \rangle$ to denote a phase consisting of ions whose separations (measured in lattice spacings) repeat periodically the sequence $n_1, n_2, \ldots, n_m$. (Hence the phases $n_i = s_i = 1$ and $n_i = s_i = 0$ can be described as $\langle 1 \rangle$ and $\langle \infty \rangle$, respectively.)

The multi-degeneracy encountered on the phase boundaries of figure 1 is due to the absence of interaction between the confined electrons. It is natural to expect that, for $t/U \neq 0$, the hopping of electrons will introduce an effective coupling between the ions, thus providing a mechanism for the removal of the degeneracy. This intuitive picture can be formalized using the defect–defect interactions introduced by Fisher and Szpilka [8]. In the present context, a defect corresponds to an ion. Following [8] the energy per lattice site of phase $\langle n_1, n_2, \ldots, n_m \rangle$ can be written as $E_{\langle n_1,\ldots,n_m \rangle} = E_{\mathrm{tot}} / \sum_{i=1}^{m} n_i$, where

$$E_{\mathrm{tot}} = m\sigma + \sum_{i=1}^{m} V_2(n_i) + \sum_{i=1}^{m} V_3(n_i, n_{i+1}) + \cdots \tag{3}$$

and σ is the creation energy of an isolated ion, $V_2(x)$ denotes the effective interaction between two ions at a distance x, $V_3(x, y)$ the interaction of three ions with spacings x, y, and so on. Although, for simplicity, we refer to the ion creation energy and ion–ion interactions, it must be borne in mind that each ion is associated with an electron.

When $t = 0$ the electrons are confined to the ions. In this case σ is readily shown to be equal to $-2h$. For small t/U we expect each electron to be localized in a region around the associated ion. Using standard perturbation theory one can obtain σ to leading order in t/U

$$\sigma = -2h - 2t^2/U + \mathcal{O}(t^4/U^3). \tag{4}$$

The leading-order corrections to σ are associated with a virtual process in which the electron hops to the site to the immediate right (or left) of the ion and back again.

The general ion–ion interaction term, $V_m(n_1, n_2, \ldots, n_{m-1})$, can be obtained, at least in principle, through a reconnection formula [9]. In terms of the four different ionic configurations shown in figure 2, this formula is

$$V_m(n_1, n_2, \ldots, n_{m-1}) = E_{\mathrm{A}} - E_{\mathrm{B}} - E_{\mathrm{C}} + E_{\mathrm{D}}. \tag{5}$$

In the absence of electron hopping, equation (5) gives $V_m = 0$ for all values of m. Our aim is to calculate V_m to leading order in t/U.

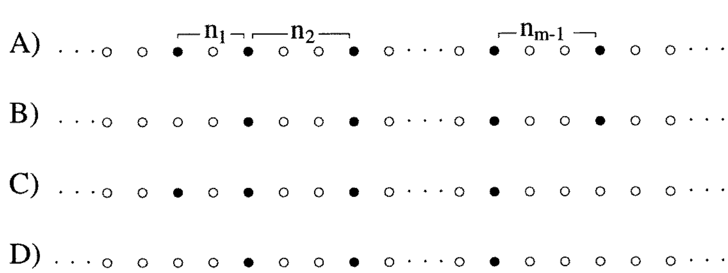

Figure 2. Ionic configurations needed to calculate the m-ion interaction $V_m(n_1, n_2, \ldots, n_{m-1})$. In A there are m ions with successive separations $n_1, n_2, \ldots, n_{m-1}$. In B the leftmost ion is removed; in C the rightmost ion is removed; and in D both the leftmost and rightmost ions are missing.

We now illustrate how this method can be used to give a result for $V_2(n)$. To calculate E_A in equation (5) consider a system of $n+1$ sites with ions at sites 0 and n. The single-particle energies are determined by the eigenvalues of the $(n+1)$-dimensional matrix, $\mathbf{M}$, where

$$M_{ij} = -U\delta_{i,j}(\delta_{i,0} + \delta_{i,n}) + t(\delta_{i,j+1} + \delta_{i,j-1}) \tag{6}$$

and the other matrix elements are zero. Two of these energies occur near $-U$, and these are the ones we want to sum over. So we write

$$E_A = \frac{1}{2\pi \mathrm{i}} \int_\Gamma \mathrm{Tr}[(z\mathcal{I} - \mathbf{M})^{-1}] z \, \mathrm{d}z \tag{7}$$

where the contour Γ encloses the region near $z = -U$ and $\mathcal{I}$ is the identity matrix. To evaluate the trace we expand the matrix inverse in equation (7) in powers of the t's. Define a perturbation $V_{ij} = t_{ij}(\delta_{j,i+1} + \delta_{j,i-1})$. Then

$$(z\mathcal{I} - \mathbf{M})^{-1}_{ii} = G_{ii} + G_{ii}V_{ij}G_{jj}V_{ji}G_{ii} + G_{ii}V_{ij}G_{jj}V_{jk}G_{kk}V_{kl}G_{ll}V_{li}G_{ii} + \cdots \tag{8}$$

where $G_{ii} = [z - M_{ii}]^{-1}$ and we use the repeated index summation convention. We therefore have established a one-to-one correspondence between terms in the expansion of equation (8) and walks on the lattice which begin and end on the same site i, in which we associate a factor $t_{ij} = t_{ji}$ with each step between sites i and j and set $t_{ij} = t$ at the end of the calculation. Note that any walk which does not include the two end sites of the configurations in equation (5) will be eliminated by the subtractions in this equation. To see this, suppose that the walk does not include the leftmost ionic site in configuration A. Then the contribution of this walk in configuration A is the same as that in configuration B, and that in configuration C is the same as that in configuration D. Therefore, such a walk has a vanishing contribution to the right-hand side of equation (5). An equivalent formulation is to say that contributions to V_m only arise from terms in equation (8) which explicitly depend on all the t_{ij}'s in the configuration under consideration. Since we obviously only need to focus on such contributions, we will introduce the notation $[\ ]'$ to denote an expression that includes only these terms. Thus, for instance, $[E_A]'$ denotes only those contributions to E_A which involve all the t_{ij}'s in the configuration being considered.

We now evaluate the terms in equation (8) which contribute to $[E_A]'$ at lowest order in t. If i is not an end site, in order to involve all the t's the matrix elements must either (a) start at i, say, then increase to the highest number site (n), then decrease to the lowest number site (0) and finally increase back to the original value i or (b) initially decrease to 0, then increase to n, and finally decrease back to i. If $i = 0$ or n, however, note that the index can only initially increase or decrease, respectively. So to leading order the terms in E_A which survive the subtractions of equation (5) are

$$[(z\mathcal{I} - \mathbf{M})^{-1}_{ii}]' \approx C_i G_{00} G_{ii} G_{n,n} \prod_{j=1}^{n-1} G_{jj}^2 \prod_{j=0}^{n-1} V_{j,j+1}^2 \tag{9}$$

where $C_i = 1$ if $i = 0$ or $i = n$ and $C_i = 2$ otherwise. The product over G's does not include the end sites, because these, in general, only appear once. The starting site appears an extra time and gives rise to the prefactor G_{ii}. The relevant term of order t^{2n} in equation (8) is

$$[\mathrm{Tr}(z\mathcal{I} - \mathbf{M})^{-1}]' \approx t^{2n} \left[\frac{2}{(z+U)^3 z^{2n-2}} + \frac{2n-2}{(z+U)^2 z^{2n-1}} \right]. \tag{10}$$

Here the first term includes C_1 and C_{n+1}, both of which are unity. The factor $2n-2$ comes from $\sum_{i=1}^{(n-1)} C_i$. Substituting (10) into (7) and calculating the integral using residues gives

$$[E_{\mathrm{A}}]' = \frac{1}{2\pi \mathrm{i}} \int_\Gamma t^{2n} \left[\frac{2}{(z+U)^3 z^{2n-3}} + \frac{2n-2}{(z+U)^2 z^{2n-2}} \right] \mathrm{d}z = (2n-2) t^{2n}/U^{2n-1}. \tag{11}$$

Next, to use the reconnection formula (5), we need to repeat the same calculation when one of the end ions is removed (corresponding to configurations B and C in figure 2). In this case

$$[\mathrm{Tr}(\mathcal{I} - \mathbf{M})^{-1}]' = t^{2n} \left[\frac{1}{(z+U)^2 z^{2n-1}} + \frac{2n-1}{(z+U) z^{2n}} \right].$$

The perturbative contributions to V_2 are thus $[E_{\mathrm{B}}]' = [E_{\mathrm{C}}]' = -t^{2n}/U^{2n-1}$. Note than when both ions are removed there are no longer any levels near $-U$. Hence $[E_{\mathrm{D}}]' = 0$ and use of the reconnection formula (5) gives

$$V_2(n) = 2nt^{2n}/U^{2n-1} + \mathcal{O}(t^{2n+2}/U^{2n+1}) \tag{12}$$

as expected [4].

Fisher and Szpilka [8] showed that, for systems where the ion–ion interactions, V_m, decay sufficiently rapidly with the defect spacings, a knowledge of the sign and convexity of $V_2(n)$ can provide a considerable amount of qualitative information about the phase diagram of the system. Their analysis can be applied in this context since the V_m decay exponentially with the spacings of the two outermost ions (because the ion–ion interaction is mediated by the nearest-neighbour hoppings of electrons). Therefore, as a first approximation, we shall analyse the phase diagram neglecting interactions that involve more than two ions. Higher-order interactions will then be included successively to resolve the finer details of the phase structure.

In the two-ion interaction approximation the ground-state configurations correspond to equispaced electron–ion pairs. Since $V_2(n)$ is always positive and convex, as h is varied from positive to negative, n increases monotonically in steps of one lattice spacing [8], giving rise to the infinite sequence of phases

$$\langle 1 \rangle \to \langle 2 \rangle \to \cdots \to \langle \infty \rangle. \tag{13}$$

Equating the energy per site of neighbouring phases gives the position of the phase boundaries and shows that the phase $\langle n \rangle$ is stable over a region of width

$$\Delta h_n \approx \tfrac{1}{2} n\, V_2(n-1) \approx n(n-1) t^{2n-2}/U^{2n-3}. \tag{14}$$

The original multi-degeneracy is not completely lifted by $V_2(n)$ because, on the boundary between two phases, $\langle n \rangle$ and $\langle n+1 \rangle$, all mixed phases where the ions can be separated by distances n or $n+1$ are still degenerate. To determine the finer structure of the phase diagram it is necessary to consider the effect of higher-order ion interactions.

The method outlined above can be extended to calculate the m-ion interaction V_m for $m > 2$. As we shall show below, $V_m(n_1, n_2, \ldots, n_{m-1})$ depends, to leading order, only on the separation of the two outermost ions in configuration A, $n = \sum_{i=1}^{m-1} n_i$. The result is

$$V_m(n) = \frac{(2n)!}{(2m-3)!(2n-2m+3)!} \frac{t^{2n}}{U^{2n-1}}. \tag{15}$$

To prove this consider equation (8). Note that m of the diagonal elements of $(z\mathcal{I} - M)^{-1}$ are $(z+U)^{-1}$; the rest are z^{-1}. If the initial i corresponds to an ion, then a factor $(z+U)^{2m-1}$

appears in the trace; otherwise the factor is $(z+U)^{2m-2}$. In the first case there are m choices for i; two at the end with $C_i = 1$ and $m-2$ in the interior with $C_i = 2$. Thus

$$\mathrm{Tr}[(z\mathcal{I} - \mathbf{M})^{-1}]' = t^{2n}\left\{\frac{(2m-2)}{(z+U)^{2m-1}z^{2n-2m+2}} + \frac{2n-2m+2}{(z+U)^{2m-2}z^{2n-2m+3}}\right\}. \tag{16}$$

Again we stress that the dependence of (16) on the position of the m ions in the chain is only through n, the distance between the two end defects. Substituting in (7) gives

$$[E_\mathrm{A}]' = \frac{(2n-2)!}{(2m-3)!(2n-2m+1)!}\frac{t^{2n}}{U^{2n-1}}. \tag{17}$$

Similarly

$$[E_\mathrm{B}]' = [E_\mathrm{C}]' = [E_\mathrm{A}]'(2m-3)/(2n-2m+2) \tag{18}$$

$$[E_\mathrm{D}]' = [E_\mathrm{B}]'(2m-4)/(2n-2m+3). \tag{19}$$

Finally, the use of the reconnection formula (5) gives for the m-ion effective interaction, V_m, the result (15). It should be pointed out that, in principle, the leading-order expression (15) could be dominated by neglected terms of higher order in t/U if n is sufficiently large (for fixed t/U) [8, 9]. However, Gruber *et al* [4] have shown that, for $m = 2$, higher-order corrections to $V_2(n)$ are dominated uniformly in n by the expression (15), provided that t is replaced by $\tilde{t} = U[\sqrt{U^2 + 4t^2} - U]/2t$. On general grounds one expects $V_m(\{n_i\})$ to be of order $\exp(-2n\xi)$ where ξ is a correlation length with $\exp(-\xi) \approx t/U$. The replacement of t by $\tilde{t}$ reflects the fact that ξ has a development as a power series in t/U. Thus, it seems plausible to expect that, upon renormalizing t in (15), their conclusion can also be extended to $m > 2$.

We now consider how higher-order ion interactions modify the phase diagram obtained in the two-ion interaction approximation. Consider first V_3. This has the effect of partially removing the multi-degeneracy on the $\langle n\rangle|\langle n+1\rangle$ boundaries by stabilizing the mixed phases $\langle n, n+1\rangle$. This happens because the energy difference, of the mixed phase relative to the pure phases, which is given by

$$\begin{aligned}(2n+1)E_{\langle n,n+1\rangle} - nE_{\langle n\rangle} - (n+1)E_{\langle n+1\rangle} &= V_3(n, n+1) + V_3(n+1, n) - V_3(n, n)\\ &\quad - V_3(n+1, n+1)\end{aligned} \tag{20}$$

is negative. To see this, note that ΔE is essentially determined by the dominant term, $-V_3(n, n) = -V_3(2n) < 0$. The mixed phase $\langle n, n+1\rangle$ has an ion density, $2/(2n+1)$, intermediate between the pure phases $\langle n\rangle$ and $\langle n+1\rangle$.

The stability of the two new boundaries appearing at this stage of approximation, namely $\langle n\rangle|\langle n, n+1\rangle$ and $\langle n, n+1\rangle|\langle n+1\rangle$ can be determined similarly by considering four-ion interaction terms. For simplicity, we consider the stability of the former phase boundary, i.e. the one between the purer phases, $\langle n\rangle$ and $\langle n, n+1\rangle$, with respect to formation of the mixed phase $\langle n, n, n+1\rangle$. In analogy with (20), we find the energy of the mixed phase relative to the purer phases to be given by

$$(3n+1)E_{\langle n,n,n+1\rangle} - nE_{\langle n\rangle} - (2n+1)E_{\langle n,n+1\rangle} \sim -V_4(n, n, n) < 0. \tag{21}$$

Again the phase boundary is unstable to the appearance of the mixed phase $\langle n, n, n+1\rangle$. Since all interaction potentials are positive, convex and exponentially decaying with the separation of the outmost ions, we can conclude that, at every stage of the construction of the phase diagram, the introduction of neglected higher-order interactions will lead to the stabilization of mixed phases of increasingly long period. Note that in this argument, we only need to invoke properties of $V_m(\{n_i\})$ for the case when each n_i is either n or $n+1$. Our claim that $V_m(\{n_i\})$ is of order $\exp(-2\sum_i n_i\xi)$ seems justified as long as the density is not too high, i.e. as long as we are discussing the hierarchical lifting of degeneracy for the phase boundary between $\langle n\rangle$ and $\langle n+1\rangle$, when n is not small.

To summarize, we have calculated the general m-ion interaction potentials in the neutral Falicov–Kimball model to leading order in t/U at zero temperature. We thereby iteratively construct the ground-state phase diagram and conclude that the ion density versus chemical potential, h, has the form of a complete devil's staircase.

Extending the strategy for the iterative construction of the phase diagram to more than one dimension is not trivial and is the focus of an ongoing investigation.

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