Electronic polarons in an extended Falicov-Kimball model

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Abstract. – We examine the one-dimensional spinless Falicov-Kimball model extended by a hybridization potential between the localized and itinerant electron states. Below half-filling we find a crossover from a mixed-valence metal to an integer-valence phase-separated state with increasing on-site Coulomb repulsion. This crossover regime is characterized by local competition between the strong- and weak-coupling behaviour, manifested by the formation of an electronic polaron liquid. A phase diagram is presented and discussed in detail.

The Falicov-Kimball model (FKM) has found widespread use as a model of the charge physics in mixed-valence (MV) materials. Although the FKM’s description of a band of itinerant $c$ electrons interacting via a repulsive contact potential $G$ with an underlying lattice of localized $f$ electrons retains its relevance \cite{1}, the great evolution of our understanding of MV systems has required modification of the model’s original form \cite{2}. The most popular such modification is the assumption of some overlap between the $c$ and $f$ wave functions: this yields the extended or quantum Falicov-Kimball model (QFKM).

The ground state of the QFKM has for many years been a matter of controversy, with two contradictory pictures common in the literature. The homogeneous solution assumes a simple renormalization of the $G = 0$ band structure by the Coulomb interaction \cite{3}; at mean-field level, this has been used to deduce the existence of a novel electronic ferroelectric MV state \cite{4}. On the other hand, the relationship of the single-impurity QFKM to the Kondo impurity \cite{5} suggests distinct strong- and weak-coupling regimes in the lattice model \cite{6,7}.

The latter scenario leads to interesting properties of the intermediate-coupling regime, specifically the formation of electronic polarons \cite{7,8}. This electronic polaron model has recently been used to explain the transport properties of certain Heusler and Ce alloys \cite{9}; more intriguingly, such polaronic structures play a pivotal role in the theory of the exotic “valence-fluctuating” superconductivity in CeCu\textsubscript{2}Ge\textsubscript{2} and CeCu\textsubscript{2}Si\textsubscript{2} \cite{10}. It is therefore of considerable interest to study the formation of electronic polarons in the QFKM.

In this letter we address this question by a non-perturbative study of the one-dimensional (1D) QFKM below half-filling. We derive an effective Hamiltonian for the occupation of the $f$ orbitals which gives a complete description of their physics. This allows us to exactly study the intermediate-coupling regime, where we find local competition between the strong- and
weak-coupling phases. This is interpreted as a gas of electronic polarons moving in a MV background. A schematic phase diagram is presented and discussed in detail.

The 1D QFKM for spinless fermions has the Hamiltonian

\[ \mathcal{H}_{\text{QFKM}} = -t \sum_{\langle i,j \rangle} c_i^\dagger c_j + \epsilon_f \sum_j n_j^f + G \sum_j n_j^c n_j^f + V \sum_j \left\{ c_j^\dagger f_j + \text{H.c.} \right\}. \]  

(1)

Some overlap between the c- and f-electron wave functions is assumed hence the on-site hybridization term \( V \ll t \). The concentration of electrons is fixed at \( n = (1/N) \sum_j \{ n_j^f + n_j^c \} \) where \( N \) is the number of sites. In general we have \( n < 1 \) due to the presence of doped impurities. We assume a MV state at \( G = 0 \), requiring the Fermi level to be pinned at

\[ -2t < \epsilon_f < -2t \cos(n\pi) \]

in the non-interacting system \([2]\).

Bosonization is an asymptotically exact method, rigorously describing the long-wavelength collective behaviour of the itinerant electrons; the short-range behaviour remains fermionic, however, and a cut-off \( \alpha \) on the wavelength of the bosonic density fluctuations is therefore imposed \([11]\). In simple cases such as the Hubbard model, \( \alpha \) can be taken as vanishing. This is not true for systems such as the QFKM, where the interactions with the localized orbitals determine the lattice constant as the minimum physical length scale. We thus assume finite \( \alpha > a \) in the QFKM \( \alpha \) parameterizes the electron delocalization length.

A finite wavelength cut-off may be easily included into the standard bosonization formalism \([11]\). In particular, for the linearized fermionic fields about the Fermi points the well-known Mandelstam representation still holds

\[ c_{\nu,j} = \sqrt{\frac{A_0}{\alpha}} \hat{F}_\nu \exp \left\{ -i \nu \left[ \phi(x_j) - \nu \theta(x_j) \right] \right\}, \]  

(2)

where \( \phi \) and \( \theta \) are the dual Bose fields, and the subscript \( \nu = L(-), R(+) \) as subscript (otherwise) for the left- and right-moving fermions, respectively. The Bose fields are defined in terms of particle-hole excitations about the two Fermi points. For a system of length \( L \gg a \) we have

\[ \phi(x_j) = -i \sum_\nu \sum_{k \neq 0} \frac{\pi}{kL} \rho_\nu(k) \Lambda_\alpha(k) e^{ikx_j}, \]  

(3)

\[ \theta(x_j) = -i \sum_\nu \sum_{k \neq 0} \nu \frac{\pi}{kL} \rho_\nu(k) \Lambda_\alpha(k) e^{ikx_j}, \]  

(4)

where \( \rho_\nu(k) = \sum_{\nu'} c_{\nu,k'}^\dagger c_{\nu',-k'} \) are the particle-hole density operators. The wavelength cut-off is enforced in eq. (3) and eq. (4) by the function \( \Lambda_\alpha(k) \): this function satisfies the conditions \( \Lambda_\alpha(k) \approx 1 \) for \( |k| < \alpha^{-1} \) and \( \Lambda_\alpha(k) \approx 0 \) otherwise. The numerical constant \( A \) in eq. (2) is determined by the functional form of \( \Lambda_\alpha(k) \). \( \hat{F}_\nu \) is the so-called Klein factor \([11]\).

The long-wavelength (\( \gg a \)) fermionic anticommutators and correlation functions are correctly reproduced by eq. (2). Since the Bose fields cannot resolve separations less than \( \alpha \), however, the Mandelstam identity breaks down at short distances \( \sim a \). This is reflected in the Bose field commutators, which are “smeared” by the cut-off function. For example, assuming exponential cut-off \( \Lambda_\alpha(k) = \exp(-|\alpha| |k|/2 \) we have \( \left[ \phi(x_j), \theta(0) \right]_- = -i \arctan(x_j/\alpha) \) and \( \left[ \partial_x \phi(x_j), \theta(0) \right]_- = -i \alpha (\alpha^2 + (x_j)^2)^{-1} \). In the limit \( \alpha \to 0 \) we recover the Luttinger model forms of a sign and Dirac delta function, respectively \([11]\).

Substituting eq. (2) into eq. (1) and applying standard field-theory techniques, the bosonized form of the QFKM Hamiltonian may be easily obtained. For an insight into the physics, however, we must rotate the Hilbert space to couple the c- and f-electron orbitals. To accomplish
this, we apply a lattice generalization of the shift transformation used by Schotte and Schotte in the X-ray edge problem, \(\hat{U} = \exp \left[ i \frac{Ga}{\pi v_F} \sum_j (n^f_j - \frac{1}{2}) \theta(x_j) \right] \) [12]. The Fermi velocity is defined \(v_F = 2ta \sin(k_F a)\), where \(k_F = n_0^c \pi/a\), with \(n_0^c\) the non-interacting c-electron concentration.

In the transformed Hamiltonian we combine the \(f\)-electron operators and the Klein factors into pseudospins using a generalized Jordan-Wigner transformation \(\tau^z_j = f^+_j \hat{F}_j e^{-iv_G x_j / 2a} \exp \left[ -i \frac{\pi}{2} \sum_{j',j} \sgn(x_{j'} - x_j) \left( n^f_{j',\sigma} - \frac{1}{2} \right) \right]\). Note the relationship of \(\tau^z_j\) to the occupation of the \(f\)-orbital at site \(j\). We write the transformed Hamiltonian in its final form,

\[
\hat{U}^\dagger \mathcal{H}_{\text{FKM}} \hat{U} = \frac{v_F a}{2\pi} \sum_j \left\{ (\partial_x \phi(x_j))^2 + (\partial_x \theta(x_j))^2 \right\} + G \left( n_0^c - \frac{1}{2} \right) \sum_j \tau^z_j - \frac{G^2 a^2}{2\pi^2 v_F} \sum_{j,j'} \tau^z_j \{ i [\partial_x \phi(x_j), \theta(x_{j'})]_-. \} \tau^z_{j'} - \frac{2GA\alpha}{\alpha} \sum_j \tau^z_j \cos \left( 2 \left\{ \phi(x_j) - \mathcal{K}_\alpha(x_j) - \left[ k_F + \frac{\pi}{2a} \right] x_j \right\} \right) + 2V \sqrt{\frac{\alpha a}{\alpha}} \sum_j \left\{ \tau^z_j e^{i(1 - \frac{\alpha a}{\pi v_F})} \theta(x_j) \times \cos \left( \phi(x_j) - \mathcal{K}_\alpha(x_j) - \left[ k_F + \frac{\pi}{2a} \right] x_j \right) + \text{H.c.} \right\},
\]

where \(\mathcal{K}_\alpha(x_j) = S_\alpha(x_j) + L_\alpha(x_j), S_\alpha(x_j) = \sum_{n=1}^{\infty} \left\{ i [\theta(x_{j+n}), \phi(x_j)]_- - \frac{\pi}{2} \right\} (\tau^z_{j+n} - \tau^z_{j-n+1})\) and \(L_\alpha(x_j) = \left( \frac{Ga}{\pi v_F} - 1 \right) \sum_n \left\{ i [\theta(x_{j+n}), \phi(x_j)]_- \right\} (\tau^z_{j+n} - \tau^z_{j-n})\). We have kept all terms produced by the canonical transform in eq. (5).

The three terms in eq. (5) arising from the Coulomb interaction are of special note. The first term represents a renormalization of the \(f\)-level energy and drives a valence transition with increasing \(G\). For \(0 \leq n_0^c \leq 0.5\) (\(0.5 \leq n_0^c \leq 1\)) the negative (positive) sign of this energy shift implies the emptying of the \(c\)-electron \((f\)-electron\) band so that all electrons have an unambiguously \(f\)-electron \((c\)-electron\) character. The case \(0 \leq n_0^c \leq 0.5\) is particularly interesting as here the Ising interaction term orders the available \(f\) electrons into a single contiguous block, the well-known segregated (SEG) phase of the \(V = 0\) limit [13]. The origin of this interaction is the finite spread \(\alpha > a\) of the \(c\)-electron wave functions; to minimize the interorbital Coulomb repulsion this favours empty underlying \(f\) orbitals. The last term originates from the backscattering of the \(c\) electrons off the localized orbitals. In the absence of the hybridization, this will order the available \(f\) electrons into crystalline phases via a Peierls-like mechanism [14]. The addition of the hybridization, however, replaces these phases by a MV state; in the following analysis we therefore neglect the backscattering. Including this effect does not qualitatively alter our conclusions [15].

The phases of the QFKM are most usefully classified in terms of the occupation of the localized orbitals. We therefore derive an effective Hamiltonian only for the \(f\)-occupation \((i.e. \) the \(\tau\) pseudospins). This is straightforwardly achieved by replacing the Bose fields in eq. (5) with suitably chosen expectation values. At weak-coupling, Schlottmann’s renormalization group study reveals that the system flows to the \(G = 0\) fixed point [6]; at strong coupling, where the system is in the SEG phase, exact diagonalization calculations by Farkašovský in the \(V = 0\) limit indicate that the \(c\) electrons are at their non-interacting fixed point [16]. On the basis of these studies it is natural to take \(\langle \phi(x_j) \rangle = \langle \theta(x_j) \rangle = 0\) across the phase diagram.
We hence obtain the effective Hamiltonian

\[ \mathcal{H}_{\text{eff}} = -J \sum_j \tau^z_j \tau^z_{j+1} + G (n^0_0 - \frac{1}{2}) \sum_j \tau^z_j + 4V \sqrt{\frac{A_0}{\alpha}} \sum_j \tau^x_j \cos \left( K_\alpha(x_j) + \left[ k_F + \frac{\pi}{2a} \right] x_j \right). \]  

We have approximated the segregating interaction by its nearest-neighbour form where \( J = i (G^2 a^2)/\left( \pi^2 v_F \right) \left[ \partial_x \phi(a), \theta(0) \right] \). 

\( \mathcal{H}_{\text{eff}} \) is instantly recognizable as a transverse-field Ising model in a uniform longitudinal field \( h^z = G(n^0_0 - \frac{1}{2}) \). For large \( G \), we recover the SEG phase as the Ising term dominates the Hamiltonian. In the limit of vanishing Coulomb repulsion, however, the site-dependent transverse field \( h^z_j = 4V \sqrt{\frac{A_0}{\alpha}} \cos(K_\alpha(x_j) + [k_F + \frac{\pi}{2a}] x_j) \) dominates eq. (6). The pseudospins are therefore ordered along the x-axis with \( \langle \tau^x_j \rangle \approx \frac{1}{2} \text{sgn}(h^z_j) \). Translated back into the f-occupation “language” this implies that \( \langle f^j_1 c_j + \text{H.c.} \rangle \neq 0 \), with the occupation of each f-orbital being given by \( \langle n^f_\alpha \rangle \approx n^0_0 \), where \( n^0_0 \) is the f-electron concentration for \( G = 0 \). This clearly corresponds to a MV state.

The MV and SEG phases are characterized by quite contradictory behaviours of the f electrons: in the former the localized orbitals are in a superposition of their different valence states, while in the latter there is a phase separation between regions with different integer ionic valence. At coupling strengths intermediate between the MV and SEG phases, therefore, we expect to observe competition between these two distinct charge physics. For a vanishing longitudinal field eq. (6) displays an order-disorder transition; the longitudinal field however lifts the system from criticality and we instead find a crossover between the ordered and disordered phases [17]. To understand the details of the crossover regime, we examine the site-dependence of \( h^z_j \); specifically, we replace \( K_\alpha(x_j) \) by its expectation value.

The string operator \( K_\alpha(x_j) \) consists of two contributions, one measuring the short-range \( [S_\alpha(x_j)] \) and the other the long-range \( [L_\alpha(x_j)] \) ordering of the pseudospins. In the SEG phase, the short-range correlations of the pseudospins are very strong and so \( S_\alpha(x_j) \) vanishes; the importance of disorder in the crossover regime, however, means that here \( S_\alpha(x_j) \) has a random variation. In contrast, the long-range ordering of the pseudospins in the SEG phase persists deep into the crossover regime. \( L_\alpha(x_j) \) is only non-zero for \( 0 \leq n^0_0 < 0.5 \) where it takes a maximum at the edge of the f-electron block, increasing linearly as this point is approached from either side: for the boundary of the f-block at \( x_0 \) we find \( L_\alpha(x_j) \sim \left( \frac{G_0}{\pi v_F} - 1 \right) |j| \).

The linear variation of \( L_\alpha(x_j) \) implies a quasiperiodic transverse field in the effective Hamiltonian eq. (6). The periodicity of this field is in general incommensurate with the lattice. Furthermore, we have made the standard approximation [18] to neglect higher-order harmonics in the decomposition of the physical fermions into the spinor components \( c_{\alpha \sigma} \). With a more complete treatment, higher harmonics originating from the curvature of the c-electron dispersion will appear in the transverse field in eq. (6): numerical [19] and analytical [20] studies have revealed that the effect of even very weak incommensurate higher harmonics is qualitatively identical to a random variation of the transverse field. Of particular note, the wave functions of the associated Jordan-Wigner fermions undergo an extended region of spectral transitions between the “pure” FM and PM forms [19]. The physical interpretation of dilute regions of order (disorder) existing within a disordered (ordered) phase is qualitatively identical to the situation in a Griffiths regime [21]. Furthermore, throughout this region the free energy is singular, immediately suggesting a parallel to the non-analytic behaviour of thermodynamic quantities characteristic of Griffiths physics [22]. Note that the incommensurability in eq. (6) is not explicitly present in eq. (1): it results from the rotation of the Hilbert space by the canonical transform.
Fig. 1 – Ground-state phase diagram for the QFKM below half-filling. The crossover regime (CR) is found on either side of the solid line \( \delta = 0 \). Along the line \( n_0^c = 0.5 \) the CR is replaced by a quantum critical point (QCP). The dotted lines denote the approximate “boundary” of the CR with the Griffiths phases G1 and G2. The dashed lines separate these Griffiths phases from the segregated (SEG) and mixed-valence (MV) phases. Electronic polarons are present in both Griffiths phases and the CR. As inset we show the phase diagram in the \( \delta-H \) plane: see text for details.

Fisher has used a real-space renormalization group (RG) method to examine in detail the random-transverse-field Ising model \[23\]. To utilise his results we approximate the transverse field in eq. (6) by a random field that is, we study the Hamiltonian

\[
\tilde{H} = -J \sum_j \tau^z_j \tau^z_{j+1} + h^z \sum_j \tau^z_j + \sum_j \tilde{h}^x_j \tau^x_j,
\]

where \( J \) and \( h^z \) are as in eq. (6) whereas the field \( \tilde{h}^x_j \) varies randomly with \( j \), the values being drawn from a cosine distribution \( \rho(\tilde{h}) d\tilde{h} = (C \pi)^{-1} \sqrt{1 - (\tilde{h}/C)^2} \), where \( C = 4V \sqrt{\frac{\alpha}{\alpha}} \). Following Fisher we find it convenient to define the dimensionless parameter \( \delta = \log(G_c/G) \), where \( G_c^2 = kt \sin(n_0^c \pi)V \). Assuming exponential cut-off, the proportionality constant takes the form \( k = 8 \sqrt{\frac{2 \pi \alpha}{\alpha}} \left[ \frac{\alpha}{2} + \frac{\alpha}{2} \right] \). Although the dependence upon \( \alpha \) obviously restricts the predictive powers of our analysis, it does not however prevent us from drawing a schematic phase diagram: for this purpose it is acceptable to choose \( \alpha = \text{constant} \). Without loss of generality we therefore set \( k \) equal to unity, which is equivalent to renormalizing the Coulomb interaction \( G \to G/\sqrt{k} \).

The physics of \( \tilde{H} \) may be defined entirely in terms of the dimensionless parameters \( \delta \) and \( H = h^z/\Omega \), \( \Omega = \max\{J, h^x_j\} \). This gives the phase diagram presented as the inset in fig. 1. The different phases and their boundaries are explained below. The dot-dashed line is the variation of the applied field with \( \delta \), which is \( G(n_0^c - \frac{1}{2}) \sim e^{-\delta} \). The intersection of this line with the different phases allows us to draw the QFKM’s phase diagram (fig. 1) in terms of the more convenient variables \( G^2/tV \) and \( n_0^c \). We note that the phase diagram for \( 0.5 < n_0^c \leq 1 \) can be obtained by reflecting fig. 1 along the line \( n_0^c = 0.5 \). The only significant difference
to fig. 1 is that the SEG phase is replaced by the “empty” state \( n_f = 0 \). We obtain the \( V \rightarrow 0 \) weak-coupling crystalline phases in the phase diagram on the inclusion of the backscattering corrections in eq. (5). Physically, since a finite hybridization is always present, a MV state will be realized instead of these phases, as we have observed above. Below we summarize the physical properties of the different phases as revealed by Fisher’s RG analysis. For the details of the relevant (but lengthy) calculations, the reader is referred to the original material [23].

Starting in the segregated phase \( (\delta \ll 0) \) we lower \( G \). Below the value \( G = \sqrt{\pi/2} G_c \) \( (\delta \approx -0.452) \), the Ising coupling in eq. (6) is no longer greater than the transverse field everywhere on the lattice. The rare regions where the transverse-field term is strongest breaks the single \( f \)-electron block up into randomly distributed large clusters separated by regions of the MV phase: within our approximation, this local competition between the MV and SEG phases may be regarded as a Griffiths phase (G1 in fig. 1) [23]. The system still, however, retains recognizable features of the SEG phase: the mean \( f\rightarrow f \) correlations \( \langle n_f^j n_f^{j+e} \rangle \) decay exponentially towards \( (n_f^f)^2 > (n_f^c)^2 \) with a correlation length \( \xi \sim e^{-2\delta^2} \), the non-universal power law dependence reflecting the presence of the infinite \( f \)-cluster throughout G1.

The renormalization of the \( f \)-level by the Coulomb interaction acts like a uniform longitudinal field \( h^z \), lifting the system from criticality at \( \delta = 0 \). In Fisher’s analysis, the phase diagram for finite \( h^z \) is identical to that at finite temperature: as \( G \) is lowered, the \( T = 0 \) Griffiths phase G1 gives way to a crossover regime (CR) dominated by the physics of the \( h^z = 0 \) quantum critical point (QCP). The CR is distinguished from G1 by the coexistence at every length scale of equal regions of mixed- and integer-valence. Due to the \( f \)-level renormalization we do not find algebraic decay of correlation functions, but rather exponential attenuation with a correlation length of \( \xi \sim 1 + \mathcal{O}(\delta^2) \). For \( n_0^c = 0.5 \) the \( f \)-level shifting vanishes and so the CR shrinks down to the QCP, characterized by dynamical critical exponent \( z = \infty \) at \( G = G_c(n_0^c = 0.5) = \sqrt{IV} \). Note that the precise position of the QCP is dependent upon the choice of \( \alpha \) [15]. The boundary of the CR in the inset to fig. 1 has been calculated using Fisher’s estimate \( |\delta \ln(1/H)| = 1 \) [23].

Further decreasing \( G \) below the CR, we reach a second Griffiths phase (G2) with opposite character to G1: here the hybridization term dominates the segregating interaction over most of the lattice. The SEG phase almost completely disappears, with only rare clusters of integer valence \( f \) orbitals embedded in an MV background remaining of this state. The mean correlation functions decay exponentially, although less rapidly than in G1, with \( \xi \sim (1 + \delta^{-2})^{-1} \).

Since the minimum magnitude that the transverse field in \( \mathcal{H}_{\text{eff}} \) assumes is \( \min |\tilde{h}^z_T| = 0 \), the weakly disordered phase should be present for any \( G > 0 \); for \( \delta \gg 1 \) the integer-valence clusters are however extremely rare and the last two terms of \( \mathcal{H}_{\text{eff}} \) dictate the physics. As such, we identify the very low-\( G \) behaviour as typical of a MV state (fig. 1). The MV boundary in fig. 1 is schematic, corresponding to \( \delta \approx 0.693 \).

The characteristic feature of the Griffiths phases and the CR is the local competition between the MV and SEG states. This is manifested as the co-existing clusters of \( f \) orbitals with mixed and integer valence. The integer-valence clusters are of greatest interest: these correspond to the dressing of a \( c \)-electron by a “cloud” of near-empty \( f \) orbitals. This coupling of the \( c \)- and \( f \)-electron densities originates from the forward scattering, and constitutes an electronic polaron [8]. Electronic polarons form only at intermediate coupling: at weak coupling the screening clouds are heavily suppressed by the resonant (MV) scattering with the \( f \) orbitals whereas at strong coupling the clouds merge to form the SEG phase. Since the bosonization method treats the forward scattering exactly, our discovery of electronic polarons within a crossover regime between mixed- and integer-valence phases rigorously confirms the scenario proposed by Liu and Ho [7].
Note that in the Griffiths phases which flank the sides of the CR the length scale $\xi$ differs from the conduction electron mean free path which gives rise to competing time scales: slow motion of the electronic polarons and fast motion of the conduction electrons. The different dynamics of the two types of particles provides a close analogy to a two-fluid scenario. Since the polarons are randomly distributed across the lattice, these states can be viewed as intrinsic inhomogeneities involving charge fluctuations and short-range charge correlations. This resembles very closely the spin polaron liquid found in the 1D Kondo lattice model [24].

Summarizing our results, we have derived an effective model for the occupation of the localized orbitals in the 1D QFKM. This model predicts a crossover from a MV state at weak $c$-$f$ Coulomb repulsion $G$ to the integer-valence SEG state with increasing coupling strength. At intermediate values of the coupling the system exhibits electronic polaron effects.

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