

Phase separation in the combined Falicov-Kimball and static Holstein model

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The Falicov-Kimball model has been known to phase separate when the correlation strength is large and the temperature is low. We examine the stability of phase separation under the influence of an additional electron-phonon interaction that tries to force the electron density to be homogeneous. We find that the electron-phonon interaction eventually destroys the phase-separated state once it becomes larger than some critical value. The results of this work may have an influence on theories of colossal magnetoresistance materials.

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The phenomenon of strong electron correlations driving phase separation has been long conjectured in the Falicov-Kimball model¹ and in the Hubbard model.² Little progress has been made on rigorously establishing phase separation in the Hubbard model, but it has been proved to exist in the Falicov-Kimball model in one dimension,³ in infinite dimensions,⁴ and in all other dimensions.^{5,6} In spite of these rigorous results, there has still been only limited calculations of phase diagrams and the transition temperature as a function of the parameters in the model. Essentially all finite-temperature calculations have been performed in the infinite-dimensional limit.^{4,7-9} Here we continue with this tradition, and illustrate how competing (finite) interactions can destroy the phase-separated state, even if the Falicov-Kimball interaction strength is infinitely large.

The system we choose to study is the harmonic version of the spinless Falicov-Kimball-static-Holstein model.^{10,11} The Hamiltonian is

$$H = -\frac{t^*}{2\sqrt{d}} \sum_{\langle i,j \rangle} c_i^\dagger c_j + (E_f - \mu) \sum_i f_i^\dagger f_i + U \sum_i c_i^\dagger c_i f_i^\dagger f_i + \frac{1}{2} \kappa \sum_i x_i^2 + \sum_i (gx_i - \mu)(c_i^\dagger c_i - \rho_e), \quad (1)$$

where c_i^\dagger (c_i) is the creation (annihilation) operator for an itinerant electron at site i , f_i^\dagger (f_i) is the creation (annihilation) operator for a localized electron at site i , $t^*/2\sqrt{d}$ is the hopping integral between nearest-neighbor sites (each pair is summed over twice for Hermiticity), E_f is the local f -electron site energy, U is the Falicov-Kimball interaction, x_i is the static phonon coordinate, κ is the spring constant (chosen to be 1 here), and g is the electron-phonon interaction. The symbol ρ_e denotes the average itinerant-electron concentration $\langle c^\dagger c \rangle$. We examine the system in the infinite-dimensional limit¹² and take t^* as our energy unit. A common chemical potential μ is used to fix the total electron concentration (itinerant plus localized). Note that we have chosen the phonons to interact with the local charge fluctuation, which is a common choice, but not the only one that

could be made. Since the term that is subtracted depends on the localized electron concentration it is not just a constant that can be included or removed; it has a nontrivial effect on the value of the Helmholtz free energy, and thereby on the phase separation. For concreteness we solve our system of equations on an infinite-coordination Bethe lattice, which has a semicircular electronic density of states

$$\rho(\epsilon) = \frac{1}{2\pi} \sqrt{4 - \epsilon^2}. \quad (2)$$

The system is solved by employing dynamical mean-field theory. The procedure simply combines the strategy used for the Falicov-Kimball model¹³ with that used for the static Holstein model.^{14,15} We summarize the procedure here to standardize our notation. Our interest is in the $U \rightarrow \infty$ limit. In that case, the itinerant electrons and the localized electrons are not allowed to sit at the same lattice site. The dynamical mean-field theory is straightforward to evaluate. The partition function becomes

$$Z = \int_{-\infty}^{\infty} dx 2e^{-(\beta/2)(gx - \mu)} \prod_n \frac{G_0^{-1}(i\omega_n) - gx}{i\omega_n} \times \exp\{-\beta[-(gx - \mu)\rho_e + \frac{1}{2}\kappa x^2]\} + \int_{-\infty}^{\infty} dx 2e^{-(\beta/2)(gx - \mu + U)} \prod_n \frac{G_0^{-1}(i\omega_n) - gx - U}{i\omega_n} \times \exp\{-\beta[-(gx - \mu)\rho_e + E_f - \mu + \frac{1}{2}\kappa x^2]\}, \quad (3)$$

with $\beta = 1/T$ the inverse temperature, $G_0(i\omega_n) = 1/(i\omega_n + \mu - \lambda_n)$ the effective medium, $i\omega_n = i\pi T(2n + 1)$ the fermionic Matsubara frequency, and $\lambda(i\omega_n) = \lambda_n$ the dynamical mean field. Once the partition function is determined (in the limit $U \rightarrow \infty$), then one can calculate the single-particle electronic Green's function via

$$G(i\omega_n) = G_n = \int_{-\infty}^{\infty} dx \frac{w_0(x)}{G_0^{-1}(i\omega_n) - gx}, \quad (4)$$

with $w_0(x)$ the phonon distribution function (when no localized electron occupies the lattice site), which satisfies

$$w_0(x) = 2e^{-(\beta/2)(gx-\mu)} \prod_n \frac{G_0^{-1}(i\omega_n) - gx}{i\omega_n} \times \exp\{-\beta[-(gx-\mu)\rho_e + \frac{1}{2}\kappa x^2]\}/Z; \quad (5)$$

the integral of $w_0(x)$ satisfies a sum rule

$$1 - \rho_f = \int_{-\infty}^{\infty} dx w_0(x). \quad (6)$$

[The sum rule is not one, because we have taken the limit $U \rightarrow \infty$. If U is finite, we would have a second phonon distribution function $w_1(x)$ to consider when calculating the Green's function, which enters when the site is occupied by a localized electron. In the $U = \infty$ limit, we can always rewrite $w_0(x) = P(x) - w_1(x)$ where the integral of $P(x)$ is 1 and can be interpreted as the $U = \infty$ phonon distribution function, and $w_1(x)$ integrates to the localized electron concentration. But in this work, we never distinguish these objects, so there is no need to elaborate further on them.] To complete the loop for the self-consistent solution of the problem, we need to note that Dyson's equation is

$$G_0^{-1}(i\omega_n) = G_n^{-1} + \Sigma_n, \quad (7)$$

and

$$G_n = \int_{-2}^2 d\epsilon \rho(\epsilon) \frac{1}{i\omega_n + \mu - \Sigma_n - \epsilon}, \quad (8)$$

with $\Sigma(i\omega_n) = \Sigma_n$ the itinerant-electron self-energy. This provides a closed set of equations that is needed to solve the many-body problem.

The computational algorithm that we follow is the iterative procedure introduced by Jarrell:¹⁶ (i) we begin with the self-energy set equal to zero; (ii) Eq. (8) is used to determine G_n ; (iii) Eq. (7) is used to extract the effective medium G_0 ; (iv) Eqs. (3), (5), and (4) then determine G_n ; and (v) Eq. (7) is employed to extract the new self-energy. Then steps (ii)–(v) are repeated until convergence is reached. Once the equations have converged on the imaginary axis, one can take the function $w_0(x)$ and the analytic continuation of Eqs. (8), (7), and (4) to determine the Green's function on the real axis as well.

In order to study phase separation, we also need to calculate the free energy. This is accomplished by following the procedure of Brandt and Mielsch.⁷ The Helmholtz free energy satisfies

$$F = -T \left[\ln Z + \sum_n \ln G_n - \sum_n \int_{-2}^2 d\epsilon \rho(\epsilon) \ln \frac{1}{i\omega_n + \mu - \Sigma_n - \epsilon} \right] - (E_f - \mu) \rho_f. \quad (9)$$

It is important to include a sufficient number of Matsubara frequencies in the summation to achieve convergence of the summation; indeed this ultimately limits the accuracy of our results.

It is well known that if both ρ_e and ρ_f lie between 0 and 1, then the ground state is phase separated.⁴ The key point in this work is that we fix E_f . If the chemical potential lies below E_f then the ground state contains only itinerant electrons and is not phase separated. As the chemical potential is increased, we reach a critical value of μ where the average localized electron filling becomes nonzero, and the system goes into a phase-separated state. It is straightforward to determine the value of this critical chemical potential when $g = 0$. The interacting density of states is just the semicircle described in Eq. (2) when there are no localized electrons and it becomes

$$\rho_{int}(\epsilon) = \frac{1}{2\pi} \sqrt{4(1 - \rho_f) - \epsilon^2}, \quad (10)$$

when there are ρ_f interacting electrons. So we must compare the energy with no localized electrons $E_{gs} = -(4 - \mu^2)^{3/2}/6\pi$ to the mixture of states with all localized particles and no itinerant particles (with weight ρ_f) and the state with no localized particles but with an electron filling of $\rho_e = \rho_{tot} - (1 - \rho_{tot})\rho_f$ (with weight $1 - \rho_f$) in the limit where ρ_f approaches zero. Note that one must adjust the chemical potential μ^* of the mixed state, to have the correct electron density. After performing some straightforward algebra, we find a transcendental equation for the critical value of the chemical potential,

$$E_f = -\frac{(4 - \mu^2)^{3/2}}{6\pi} - \mu \left(-\frac{1}{2} + \frac{1}{\pi} \sin^{-1} \frac{\mu}{2} + \frac{\mu}{4\pi} \sqrt{4 - \mu^2} \right). \quad (11)$$

As E_f ranges from -2 to 0 , the critical total electron density for phase separation ranges from 0 to 1 ; for $E_f > 0$ the system does not phase separate as $T \rightarrow 0$.

We will examine the case with $E_f = -0.5$ here. Equation (11) is solved by $\mu = -0.145$ and $\rho_e + \rho_f = 0.454$ for the lower boundary where phase separation begins. Hence phase separation enters when $\rho_{tot} > 0.454$. To determine the phase separation, we must perform a Maxwell construction. If we plot the electron filling as a function of the chemical potential, we will find a multivalued function in the region of phase separation. One way to perform the Maxwell construction is by drawing a line that has equal areas above and below, with the points of intersection corresponding to the densities of the phases that compose the mixture of the phase-separated state (the other way is to construct the convex hull of the free-energy curve; both methods yield the same results). The double-valued function and the Maxwell construction are plotted for $g = 1.2$ and $T = 0.04$ in Fig. 1. It is nontrivial to generate such a curve. The technical difficulty arises from the fact that for a fixed value of E_f , there are multiple solutions of ρ_f which have the same ρ_{tot} when one is in the phase separated region. Hence, we sometimes need to either adjust ρ_f and calculate E_f once the equations have converged, or employ a two-dimensional root finder for μ and ρ_f to get the equations to converge. Even so, we find regions of parameter space where we are unable to stabilize the algorithm. Note that there is an alternate way to avoid the double valuedness,¹⁷ which involves artificially shifting the

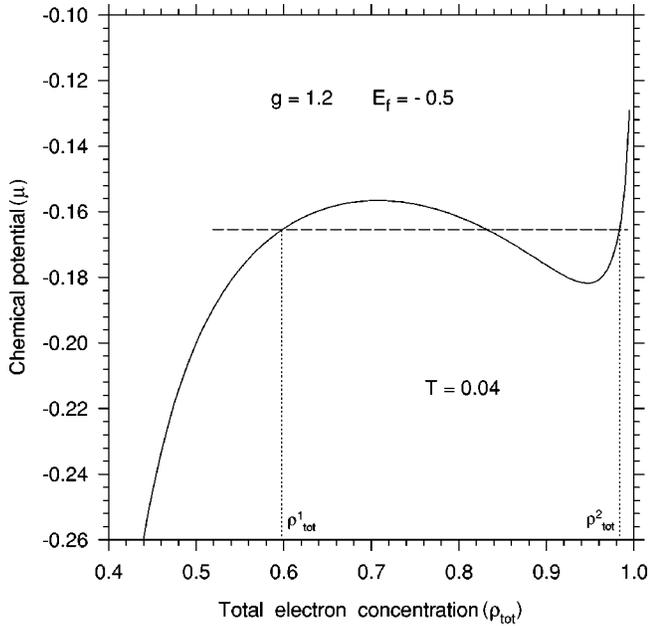


FIG. 1. The chemical potential is plotted versus the total electron filling when $g = 1.2$, $E_f = -0.5$, and $T = 0.04$. Note the double-valued chemical potential indicating phase separation. The Maxwell construction is also shown in the figure by the dashed line. The total area of the solid curve above and below the dashed line is equal to zero. The dotted lines indicate the total electron densities of the states that form the mixture.

chemical potential by $-V\rho_e$ (for some large positive constant V) to restore single valuedness. We do not employ that technique here.

We examine the transition temperature for different electron-phonon coupling strengths in Fig. 2. The phase-separated region hardly changes at zero temperature as the electron-phonon coupling increases, but the transition temperatures are sharply reduced. We run into numerical problems with the stability of our iterative solutions when g is increased to 1.2 and above. We are unable to find any strategy to solve for the phase separation at low temperature in this region of parameter space. We expect, however, that the phase separation continues to be suppressed, and conjecture that there is a critical value of the electron-phonon coupling where the phase separation disappears, but we cannot determine its value with our numerical algorithms. In addition to the binodal (first-order) phase-transition temperature, we also include the spinodal decomposition temperature (below which the system cannot be supercooled) for the $g=0$ case. This is found by determining where the $k=0$ charge susceptibility diverges. Of course the binodal and spinodal temperatures must agree at the peak of the curve.

The disappearance of the phase separation occurs when the electron-phonon interaction energy becomes similar in magnitude to the kinetic-energy gain of the phase-separated state. This occurs at a finite value of g because the kinetic-energy gain is finite, and does not diverge as $U \rightarrow \infty$.

To understand this phenomenon further, we examine in detail the system in the low-temperature limit. We already saw that when $g=0$ there is a minimum total electron den-

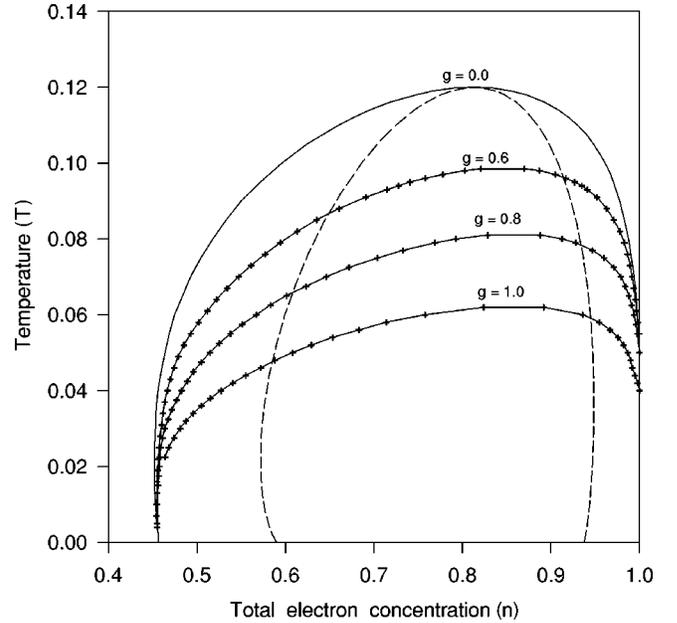


FIG. 2. The first-order phase transition as a function of the total electron filling (the solid lines show the results of the Maxwell construction). We examine four cases $g=0$, $g=0.6$, $g=0.8$, and $g=1.0$. The dashed line is the spinodal decomposition temperature (found by calculating the divergence of the $k=0$ charge susceptibility) for the $g=0$ case.

sity that must be reached before phase separation occurs. Similarly, when $g \neq 0$, we find that such a critical density continues to hold for small g . This occurs because, as T is lowered, the function $w_0(x)$ becomes sharply peaked at $x=0$, eventually becoming a delta function at $T=0$. Hence, the system remains a Fermi gas in this regime of parameter space. As g is increased to a critical value, $w_0(x)$ develops a local minimum at $x=0$ and two maxima, one at $x < 0$ and one at $x > 0$. At this point, the ground state becomes a non-Fermi liquid, and we believe that the non-Fermi-liquid character suppresses the phase separation, ultimately eliminating it. To be more quantitative, we compute

$$T \frac{d \ln w_0(x)}{dx} = g(\rho_e - \frac{1}{2}) - \kappa x - gT \sum_n \frac{1}{G_0^{-1}(i\omega_n) - gx}. \quad (12)$$

When this derivative vanishes, we have an extremum of the phonon distribution function. When $\rho_f=0$, this equation always has a solution at $x=0$ because the sum over Matsubara frequencies equals $\rho_e - 1/2$. If g is increased, we reach a point where the second derivative becomes positive, and the $x=0$ solution becomes a local minimum instead of a maximum. At this point two nonzero x solutions exist, and the system becomes a non-Fermi liquid. The second derivative (at $x=0$) is

$$T \frac{d^2 \ln w_0(x=0)}{dx^2} = -\kappa - g^2 T \sum_n G_0^2(i\omega_n), \quad (13)$$

which becomes

$$T \frac{d^2 \ln w_0(x=0)}{dx^2} = -\kappa - g^2 \int_{-2}^{\mu} d\epsilon \rho(\epsilon) \text{Re}G(\epsilon) \\ = -\kappa + g^2 \frac{(4 - \mu^2)^{3/2}}{12\pi} \quad (14)$$

in the low-temperature limit. For $\mu = -0.145$ the critical value of g to enter the non-Fermi-liquid state is 2.18.

It is useful to discuss the impact of this work on models for the colossal magnetoresistance (CMR) manganite materials. There are a number of different elements that appear to be important in describing the CMR materials. All models start with double exchange,¹⁸ but then need something else to enhance the metal-insulator transition. Three main ideas have emerged to do this: a strong electron-phonon interaction,^{14,15,19} strong disorder scattering,^{20–22} and proximity to phase separation.²³ The results of this investigation indicate that the three elements are not fully independent. Indeed, strong disorder scattering can lead to phase separation, and strong electron-phonon scattering can suppress the

phase separation. One of the consequences of this work is that one needs to carefully investigate the implications of all three of these physical effects, and how they interrelate, to construct a meaningful theory for the manganites.

In conclusion, we have analyzed the suppression of the phase-separation transition in a combined Falicov-Kimball–static-Holstein model. We see that increasing the electron-phonon coupling forces the system to be more homogeneous and suppresses the phase separation. We conjecture that a finite value of g will be sufficient to remove phase separation entirely, but are unable to access that region of the phase diagram due to numerical difficulties. Our results could have an impact on CMR materials that have both disorder and electron-phonon interactions. Any phase separation induced by the disorder may be suppressed by the electron-phonon coupling.

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