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Phase separation in the strongly correlated Falicov–Kimball model in infinite dimensions

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Abstract. Phase separation in the strongly correlated Falicov–Kimball model in infinite dimensions is examined. We show that the phase separation can occur for any values of the interaction constant J^* when the site energy ε^0 of the localized electrons is equal to zero. Electron-poor regions always have homogeneous state and electron-rich regions have chessboard state for $J^* \geq 0.03$, chessboard state or homogeneous state in dependence upon temperature for $0 < J^* < 0.03$ and homogeneous state for $J^* = 0$. For $J^* = 0$ and $T = 0$, phase separation (segregation) occurs at $-1 < \varepsilon^0 < 0$. The obtained results are exact for the Bethe lattice with infinite number of the nearest neighbours.

PACS. 71.10.Fd Lattice fermion models (Hubbard model, etc.)

1 Introduction

Phase separation, as a phenomenon, is one of the main features of the strongly correlated electron systems. In particular, it is intensively investigated in the $t - J$ model and in double exchange systems.

It should be noted that the mentioned models are very complicated from the mathematical point of view, and rigorous study of phase separation in these models has enormous difficulties. As a result, we have a few contradictory calculations which are not enough anchored for the phase separation in these models (for the $t-J$ model, especially).

However, there is a non-trivial model in the frame of which phase separation can be examined exactly. Such a model is the spinless Falicov—Kimball model [1] (for a review of the exact results in this model see Ref. [2]) with Hamiltonian

$$\mathcal{H} = -\mu \sum_i d_i^\dagger d_i + (\varepsilon^0 - \mu) \sum_i f_i^\dagger f_i + U \sum_i d_i^\dagger d_i f_i^\dagger f_i + t \sum_{<i,j>} d_i^\dagger d_j, \quad (1)$$

where $d_i^\dagger (f_i^\dagger)$ is the creation operator of itinerant (localized) electron on the site with number i, U is the intraatomic interaction and t is the transfer integral between the nearest neighbours. The chemical potential μ constrains the total number of itinerant and localized electrons. The quantity ε^0 sets the site energy level of the localized electrons in relation to the middle of the itinerant electron band. It is the important parameter of the model.

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In spite of its simplicity, the Falicov–Kimball model has a series of non-trivial physical properties. First of all, this model reveals instability of homogeneous phase towards chessboard-like charge ordering at some finite temperature T_c in a d-dimensional lattice with $d \geq 2$ [3–5]. Moreover, it is found that the pure phases (homogeneous and chessboard) are unstable in regard to phase separation at some conditions, and the Falicov-Kimball system can stay in a state with different phases coexisting in different space regions [6–11].

Showing a rich variety of the physical properties, the Falicov–Kimball model is attracted because of the possibility to investigate the mentioned properties on the rigorous basis. In particular, Brandt and Mielsch [12,13] have solved exactly this model in infinite dimensions and have created the temperature phase diagram which includes the phase separation for one case of $U = 2t$ [6], *i.e.* for the case of intermediate interaction. Using the Brandt–Mielsch results, Freericks [8,9] have calculated the zero-temperature phase diagram where one can see that the phase separation region is increased on the itinerant electron concentration when the interaction energy U changes from zero to infinity.

Recently, Freericks, Gruber and Macris [14] have considered the Falicov–Kimball model in the limit of the infinite U and have shown that the system separates (segregates) into itinerant electron-rich and itinerant electron-poor space regions having the same phase state — homogeneous state. They have shown also that a state with segregation is energetically advantageous in contrast with the pure homogeneous state.

In the present paper, we examine the phase separation for the strongly correlated Falicov–Kimball in infinite

dimensions, *i.e.* for the case of $U/t \gg 1$ and $d \to \infty$, on the basis of Hamiltonian proposed in our recent work [15]:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\rm int}, \tag{2}$$

$$\mathcal{H}_0 = -\mu \sum_i X_i^{++} + (\varepsilon^0 - \mu) \sum_i X_i^{--}, \tag{3}$$

$$\mathcal{H}_{\rm int} = t \sum_{<ij>} X_i^{+0} X_j^{0+} - J \sum_{<ij>} X_i^{++} X_j^{--} \tag{4}$$

where $J = t^2/U$ and X_i^{pq} is the Hubbard $X-$operator describing the transition of the lattice site with number i from $|q\rangle$-state to $|p\rangle$-state. In the strongly correlated regime, the lattice site has only three states, *i.e.* $p, q = +, -, 0$ where the $|+\rangle$-state is associated with the state of itinerant electrons, the $|-\rangle$-state is associated with the state of the localized electrons and the $|0\rangle$-state is the state without electrons. Double occupancy of lattice site is excluded. Hamiltonian (2) can be obtained from (1) by the canonical transformation in the limit of $U/t \gg 1$.

In the case of $\varepsilon^0 = 0$, the site energy level of the localized electrons lies in the middle of the itinerant electron band, and the model (2) presents a simplified $t-J$ model in which $\downarrow$-electrons can not move.

In reference [15], we have solved exactly the model (2) in infinite dimensions and have obtained the system of the equations describing both the homogeneous state and the charge ordering (chessboard) state for the Bethe lattice with $z \to \infty$ (z is the number of nearest neighbours). In Section 2, we present the results of our study of the phase separation on the basis of the mentioned equations and show that the strongly correlated Falicov–Kimball system separates into electron-rich regions and electron-poor regions with the corresponding phase state for any values of $J^* = Jz = \text{const.}$, $z \to \infty$. The case of $J^* = 0$ ($U = \infty$) in dependence upon the site energy ε^0 is considered in detail. Some concluding remarks will be given in last section.

2 Phase separation. Bethe lattice

Here, the phase separation in the model (1) is studied with the help of the investigation of compressibility $\mathrm{d}n/\mathrm{d}\mu$ where n is the total electron concentration. Negative compressibility indicates instability of system towards phase separation. Determination of the phase separation boundary requires use of Maxwell construction.

The necessary equations for this purpose were obtained in our previous paper [15]. For the Bethe lattice ($z = \infty$) with two sublattices A and B, these equations are exact and have the form

$$n = w + \frac{1}{2}\delta w f(-\nu) + \frac{4(a^2-b^2)^2}{W^2} \times \frac{2}{\pi} \int_0^{\pi/2} \frac{\mathrm{d}t \sin^2 t \cos^2 t}{a^2 \cos^2 t + b^2 \sin^2 t} \big(f[E(t)] + f[-E(t)]\big) \tag{5}$$

where

$$w = \frac{1}{2}(w_A + w_B) \tag{6}$$

is the concentration of localized electrons and

$$\delta w = w_A - w_B \tag{7}$$

is the order parameter of the chessboard phase. The concentrations of localized electrons w_A and w_B in the A- and B-sublattices can be obtained with the help of the following equation

$$w_i = \langle X_i^{--} \rangle = \frac{1}{\exp\beta\big[\Theta_i - \frac{1}{2}J^*\delta n_+ p_i - J^*(n-2w) - \mu_{\rm r}\big] + 1} \tag{8}$$

where

$$\beta\Theta_i = -p_i \ln\big(1 + \exp\beta(\mu_{\rm r} + \nu)\big) + \frac{1}{\pi}\int_0^{\pi/2} \mathrm{d}t \left(1 + \frac{abp_i}{a^2\cos^2 t + b^2 \sin^2 t}\right) \times \Big\{\ln\big(1 + \exp\beta[\mu_{\rm r} + E(t)]\big) + \ln\big(1 + \exp\beta[\mu_{\rm r} - E(t)]\big)\Big\}, \tag{9}$$

$$\frac{1}{2}\delta n_+ = \frac{1}{2}\delta w f(-\nu) - \nu\frac{4(a^2-b^2)^2}{W^2} \times \frac{2}{\pi}\int_0^{\pi/2} \frac{\mathrm{d}t \sin^2 t \cos^2 t}{a^2\cos^2 t + b^2 \sin^2 t} \frac{f[E(t)] - f[-E(t)]}{E(t)}, \tag{10}$$

$$E(t) = \sqrt{a^2 \cos^2 t + b^2 \sin^2 t + \nu^2}, \tag{11}$$

$$a^2 = \frac{1}{8}W^2\Big(1 - w + \sqrt{(1-w)^2 - (\delta w/2)^2}\Big), \tag{12}$$

$$b^2 = \frac{1}{8}W^2\Big(1 - w - \sqrt{(1-w)^2 - (\delta w/2)^2}\Big), \tag{13}$$

$$\nu = \frac{1}{2}J^*\delta w, \qquad \mu_{\rm r} = \mu + J^* w, \tag{14}$$

$$p_i = \begin{cases} +1, & i \in A \\ -1, & i \in B \end{cases}, \tag{15}$$

$\beta = 1/T$, $f(x)$ is the Fermi-Dirac function with the renormalized chemical potential $\mu_{\rm r}$ and W is the bare bandwidth of the Bethe lattice.

The quantities a^2, b^2 and ν^2 define the energy parameters of two bands in the ordered phase. The energetical boundaries of the bottom band are given by $-\sqrt{a^2+\nu^2}$ and $-\sqrt{b^2+\nu^2}$, and the top band lies between $\sqrt{b^2+\nu^2}$ and $\sqrt{a^2+\nu^2}$. These two bands are joined into one band in the homogeneous state where $\delta w = 0$ and $b = 0$. The halfwidth of this band is equal to $a = W\sqrt{1-w}/2$. It should be marked that the widths of the considered bands depend on w and δw in the chessboard phase and on w in the homogeneous phase. When w is increased, the widths of the correlation bands are decreased. This property is

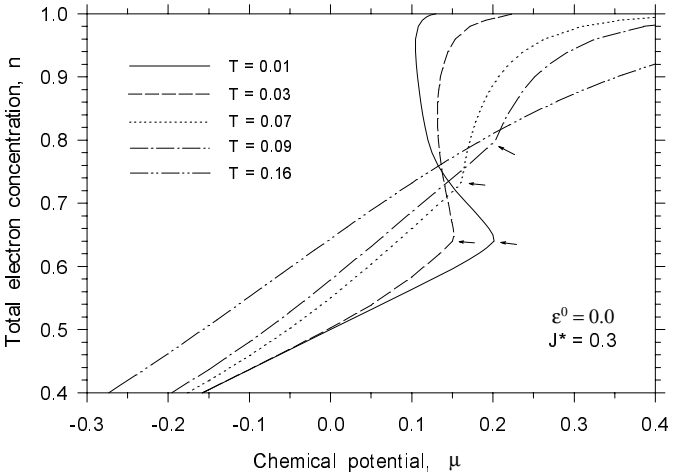

Fig. 1. Total electron concentration n as a function of chemical potential μ for $\varepsilon^0 = 0$ and different values of temperature T.

a feature of physics of the strongly correlated electron systems.

The equations (5, 6, 7) present the system of three equations for determination of the concentration of localized electrons w, the order parameter δw and the chemical potential μ for given temperature T, the total electron concentration n and the site energy ε^0. We have the equations for the homogeneous phase in the case of $\delta w = 0$. Some properties of the mentioned phases for the Bethe lattice with $z \to \infty$ were studied in reference [15].

It should be noted that these equations are written in a more simple form in contrast with reference [15].

First of all, consider the case of $\varepsilon^0 = 0$. The site energy level lies in the middle of the conduction band in this case. Figure 1 shows the chemical-potential dependence of the total electron concentration for different values of temperature T and for the case of $J^* = 0.3$ and $\varepsilon^0 = 0$ (all the energetical quantities are given in the units of $W/2$). The maximum value of the transition temperature $T_{\rm c}$ is equal to 0.15 in this case. ($n-$dependence of $T_{\rm c}$ for different values of J^* is shown in Fig. 12 of Ref. [15]). Therefore, the $\mu-$dependence of n at $T = 0.16$ is presented in Figure 1 by smooth curve. The μ-dependence of n has one break for $T < 0.15$. This break occurs at the some critical value of the total electron concentration $n_{\rm c}$ such that $\delta w = 0$ at $n \leq n_{\rm c}$ and $\delta w \neq 0$ at $n > n_{\rm c}$. The value of T marking a curve coincides with the value of $T_{\rm c}$ at $n = n_{\rm c}$ for this curve (see Fig. 12 in Ref. [15]).

We see from Figure 1 that there are curves with negative $\mathrm{d}n/\mathrm{d}\mu$ in corresponding total-concentration ranges at low temperatures. The boundaries of this ranges for every curve are determined by the values of n where $\mathrm{d}n/\mathrm{d}\mu = \infty$ or by the values of n where $n-$dependence of μ has the local maximum and the local minimum ($\mathrm{d}\mu/\mathrm{d}n = 0$). Figure 2a shows that the (n, T) phase region with the negative compressibility is arranged within the chessboard phase in the case of $J^* = 0.3$, and the part of the boundary of this region due to the local maximums coincides with the boundary of instability of the homogeneous phase.

The negative compressibility indicates instability towards phase separation and requires Maxwell construction for determination of the boundary of the region where two pure phases may be coexisted. The results of the Maxwell constructions are shown in Figure 2 by solid curves for the case of $\varepsilon^0 = 0$ and different values of J^*. A point (n, T) within a region restricted by solid curve corresponds to a state of the system where two states (two phases) coexist. These two states are defined by the points of intersection of a $T = $ const.-line passing through the point (n, T) inside the phase separation region with the solid curve. In particular, Figure 2a shows that we have two coexisting phases (homogeneous and chessboard) for the case of $J^* = 0.3$. Phase points outside the solid curve correspond to pure phases. The properties of the pure chessboard phase are shown in Figure 3 where the total-electron concentration dependence of the chessboard-order parameter δw for different values of temperature is given in the case of $J^* = 0.3$.

It is known that phase separation curve is the first order-phase-transition curve (see, for example, curve 1 in Fig. 3). Thus, the Λ-point in Figure 2a with the temperature coordinate T^* is the point where two first-order transition curves and one second-order-transition curve are converged.

It should be noticed that the general behaviour of the $\mu-$dependence of n coincides with the behaviour of the $-\mathrm{d}w/\mathrm{d}\varepsilon^0$. The poles of $\mathrm{d}n/\mathrm{d}\mu$ are the same as the poles of $-\mathrm{d}w/\mathrm{d}\varepsilon^0$. In particular, for the case of $J^* = 0.3$, the negative values of $\mathrm{d}n/\mathrm{d}\mu$ and $-\mathrm{d}w/\mathrm{d}\varepsilon^0$ are observed only at such values of n and w which correspond to the chessboard phase. In the homogeneous phase, the values of $\mathrm{d}n/\mathrm{d}\mu$ and $-\mathrm{d}w/\mathrm{d}\varepsilon^0$ are positive at $J^* = 0.3$. This is seen from the $f-f$ correlation function $\chi^{--}(\mathbf{q})$ (see Ref. [15]) taken at $\mathbf{q} = 0$. We have

$$\chi^{--}(\mathbf{q} = 0) = \frac{w(1-w)}{D(0)} \tag{16}$$

where for the Bethe lattice with $z \to \infty$

$$D(0) = T + 2w(1-w)J^*V(0) - 2w(1-w)\big[(W/4)^2 + (1-w)(J^*)^2\big]\Pi(0), \tag{17}$$

$$V(0) = \frac{1}{\pi}\int_0^\pi \mathrm{d}t f(a\cos t), \tag{18}$$

$$\Pi(0) = -\frac{1}{\pi}\int_0^\pi \mathrm{d}t \sin^2 t f'(a\cos t) = -\frac{1}{a}\frac{1}{\pi}\int_0^\pi \mathrm{d}t \cos t f(a\cos t) \tag{19}$$

where $f'(x)$ is the derivative of $f(x)$ with respect to x.

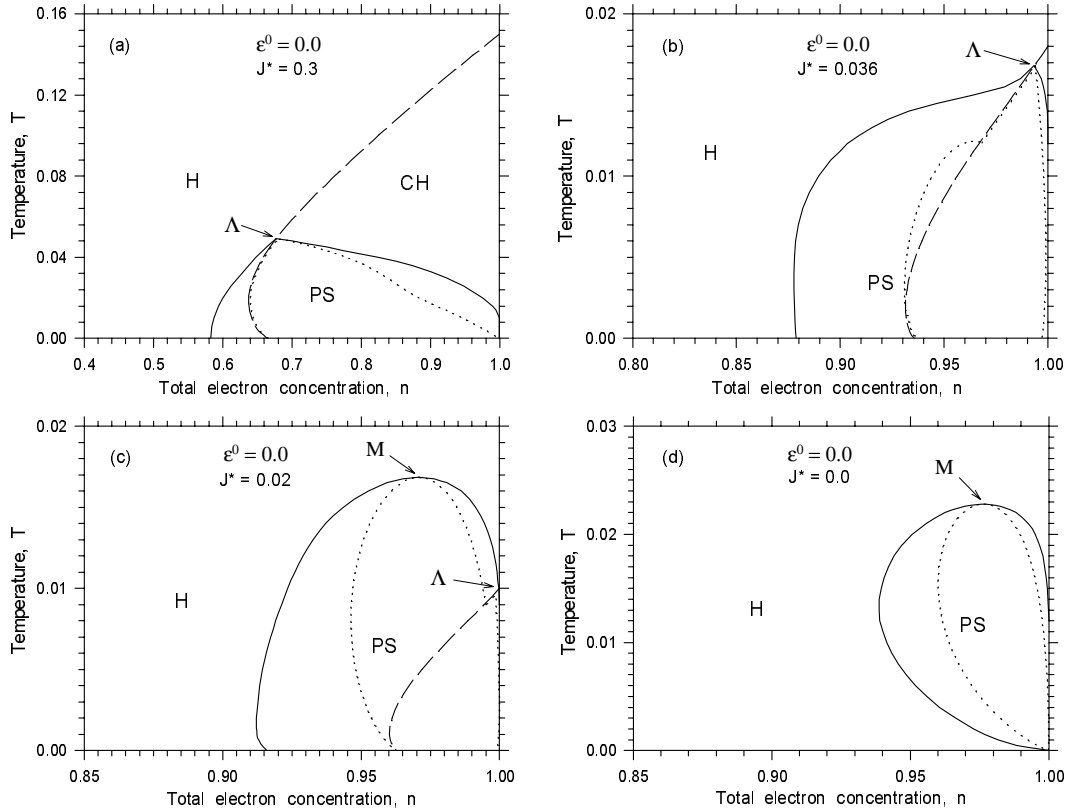

Fig. 2. Temperature phase diagram of the strongly correlated Falicov-Kimball model at $\varepsilon^0 = 0$ and different values of J^*. H, CH and PS denote homogeneous phase, chessboard phase and phase separation. Dashed curve defines the boundary of instability of homogeneous phase towards the chessboard phase, dotted curve defines region with the negative compressibility. The phase separation region is defined by solid curve.

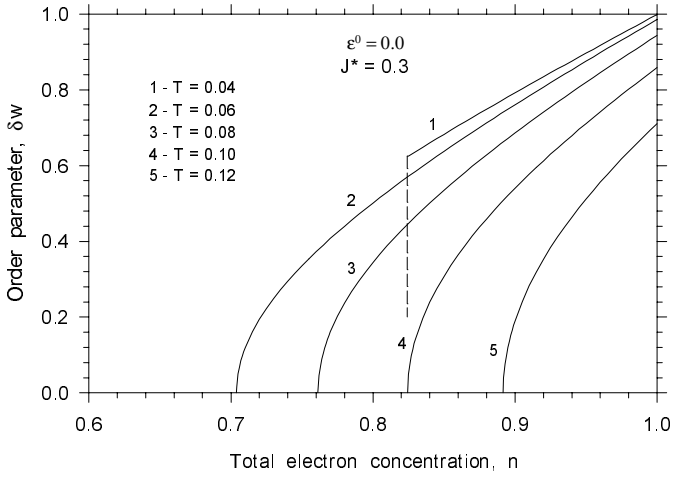

Fig. 3. Chessboard-order parameter δw as a function of total electron concentration n for $\varepsilon^0 = 0$ and different values of temperature T.

The concentration w and the chemical potential μ in the homogeneous phase are determined from the equations

$$n = w + (1-w)\frac{2}{\pi}\int_{-1}^{1} \mathrm{d}x\sqrt{1-x^2}\,f(ax), \qquad (20)$$

$$w = \Big(\exp\beta(\varepsilon^f - \mu_{\mathrm{r}}) + 1\Big)^{-1} \qquad (21)$$

where

$$\varepsilon^f = \varepsilon^0 - J^*(n-2w) + T\frac{1}{\pi}\int_0^{\pi} \mathrm{d}t \ln\big(1+\exp\beta(\mu_{\mathrm{r}} - a\cos t)\big) \qquad (22)$$

is the renormalized site energy of the localized electrons.

The analysis of (16) with the help of equations (20, 21) ($\varepsilon^0 = 0$) shows that

$$\chi^{--}(\mathbf{q}=0) = -\frac{\mathrm{d}w}{\mathrm{d}\varepsilon^0} > 0$$

for $J^* \geq J^*_{\mathrm{c1}} \simeq 0.04$ at such values of n which correspond to the homogeneous phase. Phase region with the negative compressibility oversteps phase curve of the chessboard phase for $J^* < J^*_{\mathrm{c1}}$ (see Fig. 2b), and we can have the situation when electron-rich regions have different states in dependence upon the temperature. This picture takes place at $0 < J^* < J^*_{\mathrm{c2}} \simeq 0.03$ (see Fig. 2c). The mentioned regions have the chessboard state for the low temperatures ($T < T^*$), and these regions have the homogeneous state with a corresponding value of n for $T^* < T < T_{\mathrm{m}}$ where T_{m} is the extremum (maximum or, perhaps, minimum) temperature for phase separation. (We will see subsequently that minimum temperature is also possible

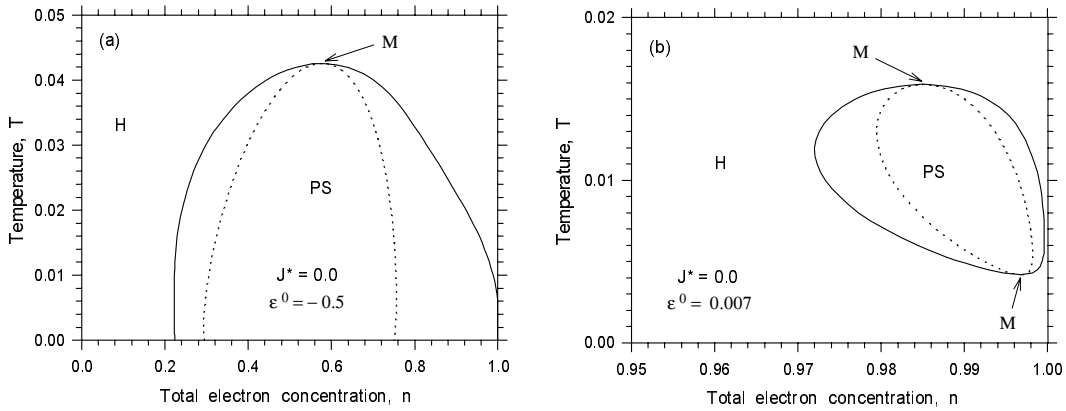

Fig. 4. Temperature phase diagram at $J^* = 0$: (a) $\varepsilon^0 = -0.5$, (b) $\varepsilon^0 = 0.007$.

for $J^* = 0$ and positive ε^0.) The electron-poor regions always have the homogeneous state for $0 < T < T_{\rm m}$. The M-point with the temperature coordinate $T_{\rm m}$ is the point where two first-order-transition curves are converged. Separation (segregation) of the system into space regions with the same homogeneous state disappears in this point.

The chessboard phase is lacking and the Λ-point is vanished in the case of $J^* = 0$ (see Fig. 2d). The system can be separated into the regions with rich electron concentration and poor electron concentration, and the phase state of these regions is homogeneous. It should be noted that the segregation region is narrowed when temperature is decreased, and the concentration range of the segregation is equal to zero at $T = 0$. This is caused by the existence of the factor $w(1-w)$ in the expression for $D(0)$ and the behaviour of w. The influence of this factor on the concentration dependence of the transition temperature $T_{\rm c}$ and the behaviour of w are discussed in reference [15]. In the case of $J^* = 0$ and $T \to 0$, the value of $w \to 0$ ($w/T \to 0$) at $0 \le n < 1$ because the chemical potential lies below the site energy level arranged in the middle of the conduction band ($\varepsilon^0 = 0$). Correlations between the itinerant electrons are absent and the segregation can not occur. When the chemical potential coincides with the site energy level, the value of w discontinuously changes from zero to 0.5. Here $n = 1$, and the n-range with the negative compressibility go to zero at $n = 1$ when $T \to 0$.

Temperature phase diagram is essentially changed for non-zero ε^0. Further, we study influence of the choice of the site energy level on phase diagram in the case of $J^* = 0$.

The site energy level lies below the middle of the conduction band for negative ε^0, and the concentration w is equal to zero for $T = 0$ at $0 < n < n_{\rm c1}$ where $n_{\rm c1} < 1$. The value of $n_{\rm c1}$ can be obtained for different values of ε^0 from Figure 8 in reference [15]. For example, $n_{\rm c1} = 0.295$ in the case of $\varepsilon^0 = -0.5$ (see Fig. 4a). Thus, the concentration range, where $\chi^{--}(0) < 0$, is limited on the left by the quantity $n_{\rm c1}$ and on the right by the quantity $n_{\rm c2}$, $n_{\rm c1} < n < n_{\rm c2}$. In the interval $n_{\rm c2} < n < 1$, the concentration w is equal to n because the chemical potential is pinned to the renormalized site energy level (the pinning of the chemical potential was discussed in Ref. [16]) and

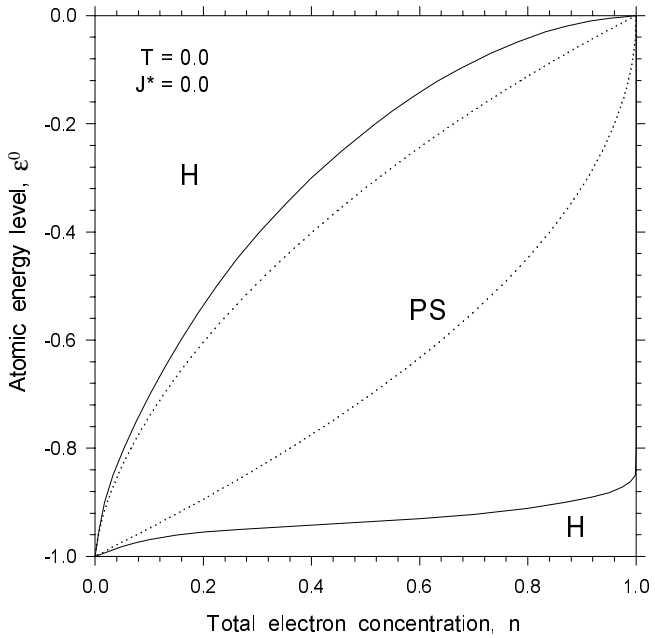

Fig. 5. Phase diagram at $J^* = 0$ and $T = 0$. The solid curve presents the separation boundary and the dotted curve determines the region with the negative compressibility.

it lies below the bottom of the conduction band which is very narrow in force of the factor $\sqrt{1-w}$ defining the width of the conduction band in the homogeneous state. $\chi^{--}(0)$ becomes the negative quantity when the chemical potential intersects the bottom of the conduction band (the quantity $\Pi(0)$ in (17) acquires a non-zero value in this case).

Figure 4b shows a case of positive value of ε^0. The site energy level lies above the middle of the conduction band in this case. One can see that the segregation is suppressed for positive values of ε^0. It is absent at any temperatures for $\varepsilon^0 > 0.0092$.

Thus, when $J^* = 0$ and $T = 0$, the separation of the strongly correlated system into electron-rich regions and electron-poor regions with the same homogeneous state for these regions occurs at $-1 < \varepsilon^0 < 0$. The (n, ε^0)-phase diagram of the considered system is shown in Figure 5

for $J^* = 0$ and $T = 0$. The space regions with different electron concentrations coexist inside of the phase region limited by solid curves. When $\varepsilon^0 \to 0$ and $\varepsilon^0 \to -1$, the separation is suppressed. The critical quantities n_{c1} and n_{c2} have the same value at $\varepsilon^0 = 0$ and at $\varepsilon^0 = -1$. $n_{c1} = n_{c2} = 1$ when $\varepsilon^0 = 0$ and $n_{c1} = n_{c2} = 0$ when $\varepsilon^0 = -1$.

3 Concluding remarks

In the present paper, we studied phase separation in the strongly correlated Falicov–Kimball model. The obtained results are exact for the Bethe lattice with $z \to \infty$. We saw that the phase separation into electron-rich regions and electron-poor regions can occur for any J^* ($\varepsilon^0 = 0$). Electron-poor regions always have the homogeneous state and electron-rich regions have the chessboard state for $J^* \geq J^*_{c2} \simeq 0.03$, the chessboard state or the homogeneous state in dependence upon temperature for $0 < J^* < J^*_{c2}$ and the homogeneous state for $J^* = 0$.

The charge ordered phase is lacking in the case of $J^* = 0$, and the system separates into electron-poor and electron-rich regions with the same homogeneous state. When $T = 0$, the segregation can occur at $-1 < \varepsilon^0 < 0$ only.

It is interesting to study influence of the choice of the site energy level on the phase separation in the case of $J^* \neq 0$. However, one can see from common considerations that the Λ-point is shifted on the chessboard-transition curve to the axis $T = 0$ for positive values of ε^0 ($T^* \to 0$) and to the axis $n = 1$ for negative values of ε^0 ($T^* \to T_c$ taken at $n = 1$). The same conclusion can be indirectly obtained from Figure 3 in reference [6]. Practical calculations must determine the critical values of ε^0 at which $T^* = 0$ (positive values of ε^0) or $T^* = T_c$ taken at $n = 1$ (negative values of ε^0) for given J^*.

It is interesting also to study space shape of the mentioned regions. In the theory of the strongly correlated systems, three possible type of shapes are discussed (for double exchange systems see, for example, Ref. [17]): stripes, spherical droplets and systems divided into two parts with different phase states. In respect to the Falicov–Kimball model one can say that realistic calculations of the shape of separated regions depend on the object of application of the model. These calculations are still absent.

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