Thermodynamic studies of the two dimensional Falicov-Kimball model on a triangular lattice

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Abstract. Thermodynamic properties of the spinless Falicov-Kimball model are studied on a triangular lattice using numerical diagonalization technique with Monte-Carlo simulation algorithm. Discontinuous metal-insulator transition is observed at finite temperature. Unlike the case of square lattice, here we observe that the finite temperature effect is not able to smear out the discontinuous metal-insulator transition seen in the ground state. Calculation of specific heat \( C_v \) shows single and double peak structures for different values of parameters like on-site correlation strength \( U \), \( f \)-electron energy \( E_f \) and temperature.

PACS. 71.45.Lr Charge-density-wave systems – 71.30.+h Metal-insulator transitions and other electronic transitions – 64.75.Gh Phase separation and segregation in model systems

1 Introduction

The Falicov-Kimball model (FKM) [1] is one of the simplest and most successful model for the correlated electron systems. This model was introduced to describe the semiconductor-metal transition in mixed-valence compounds and rare-earth systems. Recently systems such as transition-metal dichalcogenides [2,3,4], cobaltates [5], \( \text{GdI}_2 \) [6] and its doped variant \( \text{GdI}_2 \text{H}_x \) [7] have attracted considerable attention as they exhibit remarkable cooperative phenomena, like metal-insulator transition, charge and magnetic order, excitonic instability [2] and possible non-Fermi liquid states [8]. These systems are characterized by the presence of localized and itinerant electrons confined to the two-dimensional triangular lattice.

In the FKM effective interactions are mediated by band electrons [10,11]. Therefore, the model has also been studied as a model of crystallization, for example, as a model of binary alloy. The model describes the systems exhibiting different types of phase configurations e.g. regular phase [9], phase separation [10,11,12,13] and stripe phases [14,15,16]. There are predominantly more rigorous results available for the FKM on a bipartite lattice than on non-bipartite lattices. On a bipartite lattice, one of the most important result proved by Kennedy and Lieb [10,11] is that at low enough temperature the half-filled FKM possesses a long range order, i.e., the localized electrons form a checkerboard pattern, the same as in the ground state. This result holds for arbitrary bipartite lattices in dimensions \( d \geq 2 \) and for all values of on-site Coulomb correlation strength \( U \).

Recently few interesting results are reported for the phase diagram of localized \( f \)-electrons due to the inclusion of correlated hopping \((t')\) [21,22,23,24,25,26,27] of itinerant electrons in FKM. Phase segregation, even in weak correlation limit (\( U \sim 0.5 \) or 1), is one of the most striking outcome of the generalization of the FKM on bipartite [14,28] and on non-bipartite [27] lattices.

Even though the FKM is one of the simplest models for correlated electrons, a clear picture of metal-insulator transition in FKM is still debated for many years. Metal-insulator transition strongly depends upon the choice of the \( d \)-electron density of states (DOS). There are only few exact results available on metal-insulator transition on a non-bipartite lattice [27,29,30]. In a recent work [24] we have studied the ground state properties of the Falicov-Kimball Model on a triangular lattice with correlated hopping and observed first order phase transition in the \( f(d) \)-electron occupation at particular value of \( f \)-electron energy \( E_f \). This shows that the small change in the lattice parameter by means of introducing impurity or by applying pressure one can successfully explain the valence and metal-insulator transitions. In a separate work [30] we studied the effects of Coulomb repulsion between \( f \) and \( d \)-electrons (\( U \)) and also between \( f \)-electrons (\( U_f \)) themselves in a two-fold degenerate \( f \)-level on the ground state properties. We observed first order (discontinuous) insulator-metal transition at particular values of \( U \) and \( U_f \).

A very important question, in this context arises as to what is the nature of these phase-transitions at finite temperature? Does this model again give the first order (discontinuous) insulator to metal transitions of \( f(d) \)-electron occupation with \( E_f \) and temperature? In addition it is also interesting to know the dependence of the specific heat \( C_v \) on temperature for different range of parameter values.

In this work, therefore, we study the FKM in its spinless version numerically for all ranges of interactions, and
explore the metal-insulator transition and specific heat on a triangular lattice at finite temperature.

The Hamiltonian of the system may be written as

$$H = -\sum_{\langle ij \rangle} (t_{ij} + \mu \delta_{ij}) d_i^\dagger d_j + E_f \sum_i \omega_i$$

(2)

where $h_{ij}(\omega) = -t_{ij} + (U \omega_i - \mu) \delta_{ij}$.

We set the hopping integral $t_{ij} = 1$, for nearest neighboring sites $i$ and $j$ and $t_{ij} = 0$, otherwise. The eigenvalue spectrum of the Hamiltonian $H$, for a configuration $\omega$ of $f$-electrons is calculated by numerical diagonalization on a triangular lattice of finite size with periodic boundary conditions (PBC). The average value of physical quantities is obtained by the classical Monte Carlo method using Metropolis algorithm. The details of the method can be found in our earlier papers [27,30].

We work at half-filling, i.e., $N_f + N_d = N$ where $N_f$, $N_d$ are the total number of $f$- and $d$-electrons and $N = L^2 (L = 6)$ is the total number of sites. At a finite temperature $T$, the thermodynamic quantities are determined as averages over various configurations $\omega(N_f)$ with statistical weight $P(\omega(N_f))$ given by

$$P(\omega(N_f)) = \frac{e^{-\beta F(\omega(N_f))}}{Z}$$

(3)

where the corresponding free energy is given as,

$$F(\omega) = -\frac{1}{\beta} \ln(\prod_i e^{-\beta E_f \omega_i})$$
Let a thermodynamic quantity ‘A’ have value $A(\omega)$.

The partition function is

$$Z = \prod_i \left( \sum_{\omega_i=0}^{N_j} e^{-\beta E_{f} \omega_i} \right) \prod_j \left( 1 + e^{-\beta [\lambda_j (\omega(N_f)) - \mu_j]} \right).$$

(5)

Let a thermodynamic quantity ‘A’ have value $\Lambda(\omega(N_f))$ corresponding to the configuration $\omega(N_f)$ of $N_f$-electrons, then the ensemble average of ‘A’ at temperature $T$ is obtained as

$$\langle A \rangle = \sum_{N_f} \sum_{\omega} \Lambda(\omega(N_f)) e^{-\beta F(\omega(N_f))} Z.$$  

(6)

For example the ensemble average of number of f-electrons for given values of U, $E_f$, T is obtained as

$$\langle N_f \rangle = \sum_{N_f} \sum_{\omega} N_f \omega(N_f) e^{-\beta F(\omega(N_f))} Z.$$  

(7)

The specific heat $(C_v)$ is calculated from the fluctuation-dissipation theorem (FDT). The specific heat is related through the FDT to the internal energy and defined as,

$$C_v = \frac{1}{N} \frac{1}{kT^2} (\langle E^2 \rangle - \langle E \rangle^2),$$

(9)

3 Results and discussion

3.1 Noninteracting case (U=0)

We firstly study the noninteracting case ($U = 0$). Using the method described above, we have found average value of f-electron occupation number per site $n_f = \langle n_f \rangle = \frac{\langle N_f \rangle}{N}$ as a function of $E_f$ at different temperatures. Also we have calculated specific heat $C_v$ as a function of temperature for different values of $E_f$. In Fig.1 we have shown $n_f$ as a function of $E_f$ for different temperatures and also as a function of temperature for different values of $E_f$ (shown in inset of Fig.1). In Fig.2 we show the d-electron energy spectrum corresponding to the configuration $\omega(N_f)$ with minimum internal energy.

When $E_f$ is changed upwards (starting from the bottom of conduction band), $n_f$ is found to decrease suddenly at some particular positions of $E_f$. For example at $E_f=-4.0$, $n_f$ decreases abruptly from $n_f \sim 0.98$ to $n_f \sim 0.8$, at $E_f=-1.0$, from 0.8 to 0.6 at $E_f=0.0$, from 0.6 to $\sim 0.48$ and at $E_f=2.0$, from 0.48 to $\sim 0.03$. If we look at the d-electron energy spectrum, we find d-levels at energy $E=\pm 4.0, \pm 1.0, 0.0, 2.0$; i.e. there are energy gaps in d-electron spectrum in energy range $[-4, -1, -1, 0, 0, 2]$. As $n_f+n_d=1$, the chemical potential $\mu$ is pinned at $E_f$. So, as $E_f$ is shifted upward, $\mu$ shifts upward. When $\mu$ shifts from $-4$ to $-1$, there being no d-levels in this range, $n_g$ remains constant and so should $n_f$. This is exactly what we find as shown in Fig.1. As $\mu$ reaches the energy $-1$, where there are many d-levels available, these get occupied and $n_f$ decreases abruptly. Now with the increase of temperature, d-states above chemical potential are also occupied fractionally. That is why $n_f$ decreases a bit as temperature increases. However, in contrast to the case of bipartite lattice, temperature is unable to smear out the discontinuous transition on the triangular lattice. Variation of $n_f$ with temperature at different $E_f$ is shown in the inset of Fig.1. When $E_f$ is near the bottom of the d-band, $n_f$ is near unity at $T \rightarrow 0$ and decreases slowly as $T$ is
increased. At other positions of $E_f$, $n_f$ is lesser than unity (and the system is in classical mixed-valent regime) and decreases slowly as $T$ is increased.

Shown in Fig.3 is the specific heat $C_v$ as a function of temperature for different positions of $E_f$. A broad single peak is observed in specific heat curve at all values of $E_f$ except at $E_f = -2.0$. At $E_f = -2.0$ there are two peaks (one sharp peak followed by a broad peak). At low temperature we observe that variation in $C_v$ deviates from the linear behavior of free electrons. The nature of the peaks in the specific heat curves could be understood if we compare these with the d-electron energy spectrum (shown in Fig.2) for different temperatures. The d-electron spectrum does not depend on $N_f$, $E_f$ and temperatures. The spectrum is discrete and multi-gaped. Wide gap around Fermi energy in d-electron spectrum corresponds to broad peak and small gap corresponds to sharp peak in $C_v$.

3.2 Finite interaction case ($U \neq 0$)

Let us consider the case where Coulomb correlation $U$ between d- and f-electrons is finite. Fig.4(a), (b) and (c) show ($n_f - E_f$) phase diagrams for $U=1$, 5 and 10 for temperatures 0.01, 1, 2, 3 and 4 respectively. We observe that for $U=1$, as temperature increases from 0 to 4, the valence transition (i.e. $n_f$-transition) (i) occurs at higher values of $E_f$ and (ii) becomes smoother. We observe that the transition width (the range of $E_f$ over which $n_f$ goes from 1 to 0), increases as temperature increases from 0.01 to 4.

At $U=5$ and 10 the ($n_f - E_f$)-transition at different temperatures shows behavior similar to $U=1$ for $E_f$ well below or well above the middle of the conduction band. When $E_f$ is near the middle of the conduction band, further increment in the temperature does not affect the $n_f$-transition significantly. This phenomena could be explained by the large gap in the many body states around the Fermi energy ($E_F$). Due to large gap at the Fermi energy (see Fig.7 (b) and (c) for $U=5$ and 10 respectively) even higher temperature is not sufficient to move the localized f-electrons to the higher d-states.

Fig.5(a), (b) and (c) show variation of $n_f$ with temperature for $U=1$, 5 and 10 and for different $E_f$ positions. We observe that for all values of $U$, $n_f$ approaches towards the value around 0.5 at high temperatures. At low temperatures, $n_f$ decreases slowly as $T$ increases. At other positions of $E_f$, $n_f$ is lesser than unity (and the system is in classical mixed-valent regime) and decreases slowly as $T$ is increased.

Fig. 4. ($n_f - E_f$) phase diagram for different values of temperature calculated at (a) $U = 1$, (b) $U = 5$ and (c) $U = 10$.

Fig. 5. Temperature dependence of the f-electron occupation number $n_f$ at (a) $U = 1$, (b) $U = 5$ and (c) $U = 10$ for different values of $E_f$. 

Fig. 6. Specific heat ($C_v$) as a function of temperature calculated at (a) $U=1$, (b) $U=5$ and (c) $U=10$ for different values of $E_f$.

Fig. 7. (color online) The d-electron energy spectrum for different values of temperature and $E_f$ calculated at (a) $U=1$, (b) $U=5$ and (c) $U=10$.

temperatures $n_f$ takes values in the range $[0.5, 1.0]$ depending upon the values of $U$ and $E_f$. Our results are in contrast to the results of P. Farkasovsky [32] obtained on bipartite lattice. We observed discontinuous ($n_f - E_f$) transitions with temperature. We report that even higher temperature is not sufficient to smear the discontinuous ($n_f - E_f$) transition observed in ground state spinless FKM on triangular lattice [27,30]. They observed smooth variation in $n_f$ as a function of temperature. The contrast between these two results are due to the choice of different lattices. In square (bipartite) lattice, energy spectrum of the d-electrons are symmetric about the middle of the conduction band, while in triangular (non-bipartite) lattice the d-electron energy spectrum is asymmetric about the middle of the conduction band and large number of d-electron states lie in the small regime above the middle of the conduction band.

In Fig.6(a), (b) and (c) we have shown variation of $C_v$ with temperature for $U=1$, 5 and 10 at different $E_f$ positions. In the small Coulomb correlation limit (say $U=1$) and $E_f$ at the bottom of the conduction band ($E_f=-3.0$), a single broad peak in $C_v$ is observed. Moving the f-electron energy level towards the middle of conduction band two peak pattern (one sharp followed by a broad peak) is observed. For $E_f$ at center of conduction band a sharp peak followed by a low broad peak is observed.
These different peaks can be explained by the energy spectrum of the d-electrons shown in Fig.7(a). In this figure we have shown the d-electron energy spectrum corresponding to the configuration $\omega(N_f)$ with minimum internal energy. The d-electron spectrum is multi-gaped and depends upon the temperature and $E_f$. For $U=1$, $E_f=0$ and for low temperature, a gapless spectrum is observed. At finite temperature, we observe a gap around chemical potential in d-electron spectrum. Moving $E_f$ from middle to bottom of conduction band say $E_f = -2$, a large gap is observed at low temperature. The gap disappears at higher temperatures. The small gap in d-electron spectrum leads to a sharp peak while large gap corresponds to a broad peak in $C_v$.

For $U=5$ and $E_f$ at the bottom of conduction band a broad peak in $C_v$ is observed. Width of peak is reduced by moving f-electron level towards middle of conduction band. Two broad peaks in $C_v$ at $E_f = -1.0$ are observed. A single peak is observed at $E_f=0$. At $U=10$, a single peak is observed in $C_v$ for all observed $E_f$ values. The width of the peak in $C_v$ decreases with moving $E_f$ from bottom to middle of conduction band. The critical temperature ($T_c$) at which peak occurs in $C_v$, reduces as $E_f$ moves from bottom to middle of conduction band. If we compare $T_c$ for $E_f=0$ and at different U-values we observe that $T_c$ goes on decreasing with increasing U, similar to the result observed by M. Maska [29]. These features are well explained on the basis of variation of d-electron spectrum with $E_f$ and temperature shown in Fig.7(b) and (c) for $U=5$ and 10 respectively.

In conclusion, we have studied the spinless Falicov-Kimball model on a triangular lattice at finite temperature. The nature of metal-insulator transition with temperature has been looked into in various parameter regime. The temperature dependence of specific heat ($C_v$) is also studied for different ranges of parameters like $U$, $E_f$. Discontinuous metal-insulator transition is observed even at finite temperature. This is unlike the case of bipartite lattices where it has been reported that with temperature metal-insulator transition becomes second order in nature. But here we see that the temperature can not smear the discontinuous metal-insulator transition observed in ground state on triangular lattice. Single and double peak patterns are observed in $C_v$ depending on the parameters $U$, $E_f$ and temperature. It is proposed that the various features could be explained by many body spectrum of d-electrons.

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