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Possible phase separation in square and honeycomb Hubbard model: A variational cluster study

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ABSTRACT

The VCA ground state of the 2D Hubbard model is examined for possible phase separation under hole doping manifested by spatial inhomogeneities of coexisting different electron densities at equilibrium. Phase separation is accompanied by spectral weight loss and first Brillouin zone boundary deformation. Such an instability is observed in square structures and it is absent in honeycomb lattices. To our knowledge, no previous publications have revealed relationship between a Fermi surface instability and phase separation. Our VCA calculations provide strong support for this spontaneous instability, driven by electron correlations in specific lattice geometries, proposed in our earlier publications using exact quantum cluster calculations.

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1. Introduction

One of the most challenging problems in condensed matter physics has to do with understanding the physics of strongly correlated materials. Such electronic materials, found (for example) in certain oxides and selenides, give rise to a broad range of different phenomena and display complex phase diagrams. Electronic inhomogeneities originating from strong correlations have been discovered in the cuprates and iron-based superconducting materials. These spatial inhomogeneities, also often called phase separation (PS), are present as nanoscale spatial variations of electron densities in the form of stripes [1], "checkerboard" [2] and granular structures [3,4]. Theoretical investigations of strongly correlated materials are based on numerical calculations of interacting electron lattice models and most specifically are focused on the Hubbard model [5]. Recently, various numerical methods have been employed to provide insights into the many-body physics of the model, such as exact diagonalization (ED), quantum Monte Carlo (QMC) [6], dynamical mean field theory (DMFT) [7], dynamical cluster approach (DCA) [8] and variational cluster approximation (VCA) [9].

The importance of electronic phase separations driven by electron correlations was realized in the Hubbard model by Visscher [10] in the 1970s and was systematically studied at the strong interaction limit later using the $t-J$ model [11,12]. Our recent many-body calculations in different bipartite and non-bipartite clusters on phase separation in the Hubbard model are based on exact diagonalization [13–17]. These results at some critical Coulomb interactions display a level crossing instabilities similar to phase separation transitions seen at various doping levels even close to the optimal doping [1–3]. However, in spite of high accuracy cluster calculations always are inevitably tied to some uncertainties due to size and edge effects. Macridin et al. [18,19] proposed a thermodynamical description of electronic phase transition, and the theory further points to the existence of a quantum critical point which closely resembles the level crossing point observed near phase separation instability at the cluster level in Refs. [13–15]. Aichhorn et al. [20,21] discussed the coexistence of the global antiferromagnetic and superconducting long-range orders in variational cluster calculations. However, STM experiments show strong evidence that at very low temperature, electronic states on BSCCO are inhomogeneously distributed and there is no sign of any long-range ordering [3]. Therefore, here we exclusively focus on direct effects of inhomogeneities in the 2D Hubbard model at zero temperature using VCA method as an accurate variational tool to incorporate a cluster–cluster coupling in two-

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dimensional (2D) Hubbard lattice without any assumption on possible long-range ordering. Recent discoveries suggest that phase separation is likely to occur in the vicinity of a metal–insulator transition (MIT) [22–24]. In contrast to one-dimensional systems, the 2D Hubbard model in both square and honeycomb lattices at half-filling is expected to exhibit a Mott–Hubbard MIT at finite on-site Coulomb interactions U in the paramagnetic phase at both zero and finite temperature [25–30]. In general, this transition at a finite U value might be an intrinsic property of the 2D Hubbard lattice [30]. Historically, there have been contradictory claims with regard to phase separation in the 2D Hubbard model. In early QMC work [31,32], it was concluded that there was no sign of phase separation in the range of the parameters studied. Certain analytical estimates on the 2D Hubbard model by Su [33] appear to suggest that no phase separation exists at finite temperature. Some of the limitations in these approaches may have led to such conclusions. However, more recent work, using the DCA [18], has captured a phase separated region consisting of coexisting strongly correlated metallic and Mott insulating phases.

In this work, we investigate possible phase separation in both square and honeycomb lattices from half-filling to optimal doping at zero temperature following the VCA procedure. This method is known to be accurate when electron correlations are local, which is quite likely in the case of high temperature superconductors in the absence of long-range order. We find strong evidence of phase separation (co-existence of different phases) for the square lattice in the underdoped region. However, no phase separation is found in the honeycomb lattice within a similar region of doping and Coulomb interactions. Some of the important outputs of the method, such as the one-particle excitation spectral functions and spin susceptibilities in the relevant doping region, are used to identify a possible geometry-related mechanism for phase separation. The rest of the paper is organized as follows: In the following section, we formulate (in a nutshell) the principles of exact quantum cluster calculations and present the basic methodology of the variational cluster approximation. In Section 3, we study the MIT at half-filling and phase separation effects away from half-filling in square and honeycomb structures to underline their differences observed already at the cluster level. The concluding remarks are given in Section 4.

2. Method

2.1. Hamiltonian

We use one-band Hubbard Hamiltonian $\hat{H}$:

$$\hat{H} = \sum_{\langle \boldsymbol{r},\boldsymbol{r}'\rangle\sigma} (-t\hat{c}^{+}_{\boldsymbol{r}\sigma}\,\hat{c}_{\boldsymbol{r}'\sigma} + H.c.) + U\sum_{\boldsymbol{r}} \hat{n}_{\boldsymbol{r}\uparrow}\hat{n}_{\boldsymbol{r}\downarrow}, \tag{1}$$

where $\hat{c}_{\boldsymbol{r}\sigma}$ is the annihilation operator for electrons at site $\boldsymbol{r}$ with spin projection σ and U is the on-site screened Coulomb repulsion. $\langle \boldsymbol{r},\boldsymbol{r}'\rangle$ denotes summation over the nearest neighbors. The coupling parameter t is the transfer integral between the nearest neighbors. All the energies reported here are measured in units of $t > 0$. We focus here on the case with hole doping, when the electron number per atomic site ($n = N/N_a$), *i.e.*, electron density $n < 1$.

2.2. Quantum cluster calculations

The thermodynamic and ground state properties of the Hubbard model with a large number of electrons and lattice sites cannot be calculated by exact diagonalization technique. However, full diagonalization is still possible within relatively small Hubbard clusters such as squares, 8-site 2×4 ladders, two-dimensional Betts cells and pyramids [34,35].

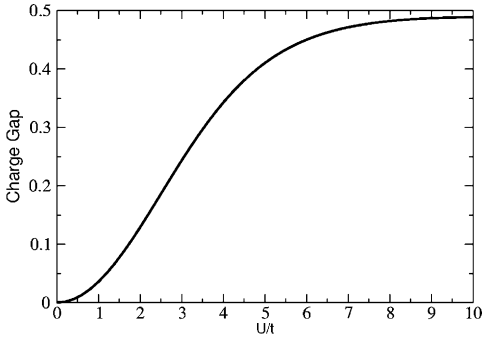

Fig. 1. The charge energy gap variation as a function of on-site interaction strength U in the 6-site honeycomb cluster. There is no sign of phase separation instability driven by U, since the gap at one hole of half-filling opens at infinitesimal U and increases monotonically.

The problem of a quantum gas of interacting many electrons in independent clusters is exactly solvable in the ground state and at finite temperatures [13–17]. The canonical and grand canonical ensembles in equilibrium display thermodynamic phase diagrams and the tendency to phase separation through responses of the system to changes of electron concentration and magnetic field. For instance, the charge excitation gap, $\Delta^c(U) = E(N+1) + E(N-1) - 2E(N)$, is calculated using the lowest canonical energies $E(N)$ with the number of electrons N in a particular doping region close to the half-filling. The positive gap region at relatively large U and one hole off half-filling describes a Mott–Hubbard insulator. The negative excitation gap $\Delta^c < 0$ earlier found in various 2×2 and 2×4 square geometries displays an electron charge instability at weak and moderate U values. The nodes of the charge gap, i.e., U values at which $\Delta^c(U) = 0$, determine the critical parameter U_c for possible level crossing instabilities. This implies a redistribution of electron density and phase (charge) separation (*i.e., segregation*) of the clusters into hole-rich and hole-poor regions for different numbers of electrons per cluster. The inhomogeneities favored by the negative gaps are essential for generating the spontaneous electron charge separation. Here we show the contrasting behavior of a charge energy gap versus U in bipartite honeycomb geometry at one hole off half-filling. In Fig. 1, the gap opens at infinitesimal U and increases with U monotonically without charge phase separation by avoiding level crossings. This positive charge gap observed at the cluster level is crucial for stabilization of homogeneous electron density in the large honeycomb lattice. Our VCA calculations in honeycomb lattice at infinitesimal U in close vicinity of half-filling confirm such insulating behavior in Fig. 1 at one hole off half-filling as $U \to 0$.

One can extrapolate the results of many-body cluster calculations obtained for small clusters to larger two-dimensional lattices and consider the "concentrated" two-dimensional Hubbard lattice modeled as a checkerboard lattice structure with embedded arrays of such disconnected unit cells, which do not interact directly, but form a grand canonical ensemble in thermodynamic equilibrium. In inhomogeneous concentrated systems, this description in thermodynamic equilibrium becomes quite accurate for suitable values of parameters since the lattice can be broken up into periodic arrays of weakly coupled clusters. In the VCA, the cluster–cluster interaction is usually added through the coupling between the unit cells comparable to the energy transfer scale t within an individual cluster. The VCA provides accurate results for cluster sizes not accessible to full diagonalization. Thus, exact predictions given by a grand canonical ensemble of clusters in various geometries can be tested using the VCA results in the "concentrated" 2D Hubbard lattice. Below we compare the geometry dependence in square and honeycomb bipartite lattices using the disconnected square and