Absence of Spin Liquid in Nonfrustrated Correlated Systems

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The question of the existence of a spin liquid state in the half-filled Hubbard model on the honeycomb (also known as graphene) lattice is revisited. The variational cluster approximation, the cluster dynamical mean field theory, and the cluster dynamical impurity approximation are applied to various cluster systems. Assuming that the spin liquid phase coincides with the Mott insulating phase in this nonfrustrated system, we find that the Mott transition is preempted by a magnetic transition occurring at a lower value of the interaction $U$, and therefore the spin liquid phase does not occur. This conclusion is obtained using clusters with two bath orbitals connected to each boundary cluster site. We argue that using a single bath orbital per boundary site is insufficient and leads to the erroneous conclusion that the system is gapped for all nonzero values of $U$.

\begin{equation}
H = -\sum_{\langle ij \rangle} (c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.}) + U\sum_{i} n_{i\uparrow} n_{i\downarrow},
\end{equation}

where $c_{i\sigma}$ annihilates a fermion of spin projection $\sigma = \uparrow, \downarrow$ at site $i$, $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ is the number of fermions of spin $\sigma$ at site $i$, and $\langle ij \rangle$ denotes the nearest-neighbor pairs on the lattice. This model attempts minimally at describing electron-electron interactions in a material like graphene, although a realistic description of graphene should involve long-range Coulomb interactions, phonons, and so on. On the other hand, systems of ultracold atoms could be arranged to be fairly accurately described by the Hamiltonian (1), with adjustable interaction strength $U$.

In a recent work on the matter, Meng et al.\textsuperscript{1}, using quantum Monte Carlo simulations, have argued that a spin liquid phase exists in model (1) in the range $3.5 < U < 4.3$. Below $U = 3.5$, the model is in a semi-metallic state, and beyond $U = 4.3$ is in an antiferromagnetic state (the two sublattices carrying opposite magnetizations). But more recently, this result has been challenged by Sorella et al.\textsuperscript{2}, also using quantum Monte Carlo simulations, albeit with larger systems. Although this controversy seems a rather technical one, rooted in Monte Carlo methods, it also shows that the model in question, if not in a spin liquid state, is very close to one.

The presence of a spin liquid phase has been supported by other works\textsuperscript{[3–5]} using quantum cluster methods\textsuperscript{6}, such as the variational cluster approximation (VCA)\textsuperscript{[7,8]}, the cluster dynamical impurity approximation (CDIA)\textsuperscript{[8,9]}, and cluster dynamical mean field theory (CDMFT)\textsuperscript{[10–13]}. Quantum cluster methods have been used extensively in the last decade to refine our understanding of the Mott-Hubbard transition and of competing orders (magnetism, superconductivity) in strongly correlated materials. They are based on some representation of the full systems by a small, finite cluster of sites, with additional uncorrelated orbitals (the “bath”) and/or adjustable source terms in the Hamiltonian. These additional elements are determined either by a self-consistency or by a variational principle\textsuperscript{[14]}. More specifically, Yu et al.\textsuperscript{[3]} have studied the question within the Kane-Mele model, which reduces to model (1) in the special case $\lambda = 0$. They support the existence of a spin liquid phase in the range $3 \leq U \leq 4$, based on the computation of the spectral function and the associated spectral gap within VCA. Wu et al.\textsuperscript{[4]} conclude similarly with CDMFT using a QMC solver. Seki and Ohta\textsuperscript{[5]} use the VCA and CDIA to study the specific question of the antiferromagnetic transition and the metal-insulator transition in model (1). They conclude that the single-particle
the presence or not of a gap may also be ascertained by computing the particle density $n(\mu)$ around the particle-hole symmetric point $\mu = U/2$ and to look for a plateau in $\mu$, which would be the signature of a gap (the constraints on $\mu$). To ascertain the presence of a spectral gap, we proceed as follows: The density of states $N(\omega)$ can be computed by numerically integrating the spectral function $A(k, \omega)$ over wave vectors. We compute $N(\omega + i\eta)$ at $\omega = 0$ for a few values of the Lorentzian broadening $\eta$ and extrapolate $\eta \to 0$ using a polynomial fit. The result of this extrapolation should vanish in the insulating solution, but not in the metallic solution. This extrapolated density of states is shown (dashed curves) in Fig. 2. The remarkable feature is that is does not vanish in the insulating solution associated with the system $s4-4b$, but does, as it should, in the large bath system $s4-8b$. Thus, even though System $s4-4b$ displays the a first-order transition that has all the appearances of a Mott transition, its spectral function in the strong-coupling phase has no gap, and thus this system does not adequately describe a Mott insulator. In the context of CDMFT or CDIA, this is related to the presence of a single bath orbital per cluster site, and to particle-hole symmetry. The latter forces the bath energy $\epsilon$ to vanish. A correct description of the insulating state requires rather a minimum of two bath orbitals per cluster site, with equal and opposite bath energies $\pm \epsilon$. The presence or not of a gap may also be ascertained by computing the particle density $n(\mu)$ around the particle-hole symmetric point $\mu = U/2$ and to look for a plateau in $\mu$, which would be the signature of a gap (the constraints on $\mu$).
Nonzero staggered magnetization $M$ coincides, as this value of $U_N$ is remarkably close to the one found in Ref. [2] ($3.75 < U_N < 3.8$). This is most likely a happy coincidence, as $U_N$ will depend on cluster size. However, the spectral gap obtained from the poles of the Green function is nonzero for all values of $U$, even those below $U_N$. This agrees with Ref. [5]. This seems to be a signature of the AF state to be fully gapped. This is observed in all systems studied (e.g., by inspecting the spectral function).

The first system studied is a six-site cluster (h6 in Fig. 1). It has been treated with VCA, using the nearest-neighbor hopping parameter $t'$ appearing in the cluster Hamiltonian $H'$ and a staggered magnetization $M$ as variational parameters (see Supplemental Material [14] for a brief summary of the method). As shown in Fig. 3, the system develops a nonzero staggered magnetization $M$ for $U > U_N = 3.75t$. This value of $U_N$ is remarkably close to the one found in Ref. [2] ($3.75 < U_N < 3.8$). This is most likely a happy coincidence, as $U_N$ will depend on cluster size. However, the spectral gap obtained from the poles of the Green function is nonzero for all values of $U$, even those below $U_N$. This agrees with Ref. [5]. This seems to be a signature of a spin liquid state, but, as we will see below, systems that are better equipped to describe the Mott transition will lead us to the opposite conclusion. Note that the gap computed from the VCA solution lies below the one obtained from the Green function without variational parameters, hinting that a better variational solution prefers a smaller gap.

System h6-6b, with one bath orbital per cluster site, was also studied, and our calculations agree with those of Ref. [5]: the system has a spectral gap for all values of $U$ and displays no Mott-insulator transition (see Supplemental Material [14]). However, we assert that probing the Mott transition in this system is unreliable, just as it is in System s4-4b for the square lattice. It is safer to use systems with two bath orbitals per cluster site. The simplest such system for the honeycomb lattice is h2-4b (Fig. 1). We studied this system both with CDMFT and CDIA. At half-filling, particle-hole symmetry demands that the bath energies of the two bath orbitals connected to the same cluster site be opposite in value ($\pm \epsilon$). In the nonmagnetic state, the two sites of the cluster (and the corresponding bath sites) are related by left-right symmetry, and therefore only two bath parameters remain: one bath energy $\epsilon$ and one hybridization parameter $\theta$. The CDMFT paramagnetic solution for $\epsilon$ is shown in the upper panel of Fig. 4. An upturn in the value of $\epsilon$ at around $U = 5.5$ signals the Mott transition. But no hysteresis is seen when the interaction $U$ is swept upwards and downwards, which means that CDMFT in this case does not detect the first-order character of the Mott transition.

Things are different when CDIA is applied to the same system. As shown again in the upper panel of Fig. 4, two CDIA solutions are found: a semi-metallic solution when $U$ is increased and an insulating solution when $U$ is decreased. Each of these stops, respectively at $U_{c1}$ and $U_{c2}$, and coexist in the range $[U_{c1}, U_{c2}]$. The first-order character of the solution is therefore clearly seen in CDIA.
Using the same extrapolating method used in the square lattice case, it is easily verified that the spectral gap vanishes throughout the semi-metallic solution, whereas it is nonzero in the insulating solution (see Fig. 5). Note that in that case, $N(\omega = 0.01i + i\eta)$, extrapolated to $\eta \to 0$ for different systems, as indicated. Only the semimetallic solutions of h2-4b and h4-6b have a non-negligible value.

FIG. 5 (color online). Top panel: Density of states in the semimetallic (SM, red) and insulating (MI, blue) solutions obtained from System h4-6b at $U = 7$, with a Lorentzian broadening $\eta = 0.05t$. Bottom panel: Density of states $N(\omega = 0.01i\eta)$, extrapolated to $\eta \to 0$ for different systems, as indicated. Only the semimetallic solutions of h2-4b and h4-6b have a non-negligible value.

The larger cluster system h4-6b (see Fig. 1) with two bath orbitals per site was also studied. In this case, two four-site clusters are necessary to form a repeated unit. Each cluster is hybridized to 6 bath orbitals, two on each edge site (the central site is not coupled to the bath). Again, particle-hole symmetry and rotational symmetry make for only two independent bath parameters, $\varepsilon$ and $\theta$, like for the smaller system h2-4b. The solution is shown on the lower panel of Fig. 4. When CDMFT is applied, the Mott transition appears clearly at $U = 6.35$, but no hysteresis is observed. Again, CDIA finds a semimetallic and an insulating (spin liquid) solution, which overlap between $U_{c1}$ and $U_{c2}$. Their energies $\Omega$ are equal at an intermediate value $U_c$ (indicated on the figure). Like in the case of the system h2-4b, the spectral gap vanishes in the semimetallic phase (Fig. 5). If the cluster Weiss field $M$ is added to the list of variational parameters, the CDIA predicts an antiferromagnetic transition at $U_N = 4.0$, which again means that the Mott transition is preempted. Thus, this larger system also rules out a spin liquid phase.

Conclusion.—We have applied various quantum cluster methods to the Hubbard model on a honeycomb lattice, in order to investigate the possible emergence of a spin liquid state. We make the hypothesis that the spin liquid state that might emerge in a strongly correlated system without magnetic frustration, such as the one studied here, coincides with the Mott insulating state. The Mott transition itself cannot be adequately accounted for by CPT or VCA: the cluster’s environment must be described by a bath of uncorrelated orbitals, i.e., by a dynamical mean field, and this bath must be large enough (two bath orbitals connected to a single cluster site). This leaves CDMFT or CDIA as adequate cluster methods to study the Mott transition. Two nonmagnetic solutions (a semimetal and an insulator, also known as a spin liquid) are found, separated by a first-order transition. The CDIA is the better approach, since it clearly reveals the three critical values $U_{c1}$, $U_{c2}$, and $U_c$. The spectral functions computed from these solutions show the persistence of the Dirac cones up to the Mott transition, hence the gapless character of the semimetallic solution. The magnetic solution can also be obtained in CDIA, and always appears at a much lower value of $U$ than the Mott transition. This leads us to assert that a spin liquid (also known as a Mott insulator) does not exist in this system: it is preempted by magnetic order. The critical value of $U$ at which the magnetic solution appears is comparable to what is found in large-scale Monte Carlo simulations [1,2]. Since the Mott transition is a rather local phenomenon, we argue that increasing the cluster size, which we cannot do with our exact diagonalization solvers, would not affect the value of $U_c$ to the point of changing our conclusion [15]. In fact, increasing the cluster size would likely shift $U_c$ to a slightly higher value. Therefore we believe that our conclusion carries over to large clusters.

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[14] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.110.096402 for (i) a brief review of the quantum cluster methods used in this work, (ii) an additional numerical argument for the metallic or insulating character of the systems studied, and (iii) sample spectral functions for the system $h_{4-6b}$.
[15] Indeed, a variational study of the Mott transition in the one-dimensional Hubbard model with NN and NNN hopping has shown that $U_c$ is rather independent of system size, even though there is a size effect in the fluctuations surrounding the transition. Note that in CDMFT and CDIA the fluctuations are temporal more than spatial, whereas temporal fluctuations are not taken into account in a variational wave function approach. See M. Capello, F. Becca, M. Fabrizio, S. Sorella, and E. Tosatti, Phys. Rev. Lett. 94, 026406 (2005).