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.

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Introduction.—There is currently a keen interest in materials and models that could display a *spin liquid* state. Such a state is characterized by the presence of local magnetic moments that do not order at any temperature; spin-spin correlations then decay exponentially as a function of distance (or algebraically, in so-called algebraic spin liquids). In theoretical language, it can be described as an insulator that is not adiabatically connected to a band insulator, but is rather a pure Mott insulator, without spontaneously broken spatial or spin symmetries.

A spin liquid could arise from the presence of strong geometric frustration, for instance in materials with a structure resembling that of the kagome lattice, such as herbertsmithite, or other kinds of frustrated geometries. It has been conjectured that spin liquids could also appear in the intermediate-coupling regime of strongly correlated systems, somewhere between a metallic (or semi-metallic) phase and a magnetic phase. It is the latter possibility that we will entertain in this Letter.

More specifically, we will address the controversy about the existence of a spin liquid in the phase diagram of the half-filled Hubbard model on the honeycomb lattice. The corresponding Hamiltonian reads

$$H = -t\sum_{\langle ij\rangle}(c^{\dagger}_{i\sigma}c_{j\sigma} + \text{H.c.}) + U\sum_{i} n_{i\uparrow}n_{i\downarrow}, \qquad (1)$$

where $c_{i\sigma}$ annihilates a fermion of spin projection $\sigma = \uparrow, \downarrow$ at site i, $n_{\sigma} \equiv c^{\dagger}_{\sigma}c_{\sigma}$ is the number of fermions of spin σ 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 Hamilonian (1), with adjustable interaction strength U.

In a recent work on the matter, Meng *et al.* [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 $U \approx 3.5$, the model is in a semi-metallic state, and beyond $U \approx 4.3$ is is in a antiferromagnetic state (the two sublattices carrying opposite magnetizations). But more recently, this result has been challenged by Sorella *et al.* [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 [3–5] using quantum cluster methods [6], such as the variational cluster approximation (VCA) [7,8], the cluster dynamical impurity approximation (CDIA) [8,9], and cluster dynamical mean field theory (CDMFT) [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 [14]. More specifically, Yu *et al.* [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 $3 \lesssim U \lesssim 4$, based on the computation of the spectral function and the associated spectral gap within VCA. Wu *et al.* [4] conclude similarly with CDMFT using a QMC solver. Seki and Ohta [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

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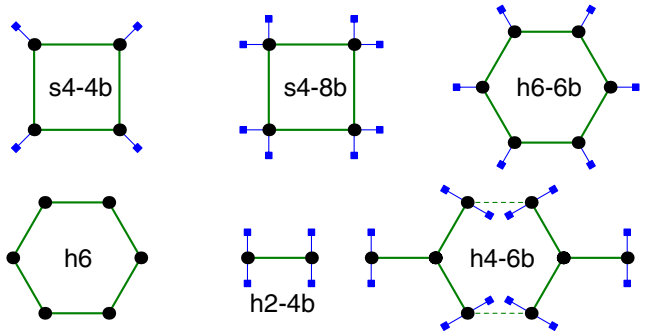

FIG. 1 (color online). Clusters used in this Letter. The first two pertain to the square lattice Hubbard model, the other to the honeycomb lattice. Blue squares represent bath sites, black circles cluster sites per se. Dashed lines represent intercluster links when more than one cluster make up the repeated unit of the super lattice.

gap opens up at an infinitesimal value of U. This also supports the existence of a spin liquid state.

In this Letter we will argue, on the contrary, that model (1) does not lead to a spin liquid phase in the intermediate coupling regime. Instead, the transition towards a spin liquid is preempted by a magnetic transition towards an antiferromagnetic state, like on the square lattice. We will also use quantum cluster methods with an exact diagonalization solver (mostly CDMFT and CDIA), except that larger bath systems will be used. Indeed, we assert that probing the Mott transition with a bath system of insufficient size may lead to the wrong conclusion.

The square and honeycomb lattice are both bipartite, and the half-filled Hubbard model defined on both lattices benefits from particle-hole symmetry, which sets the value of the chemical potential μ to $U/2$, and imposes constraints on the bath parameters used in CDMFT and CDIA. We shall therefore start with a discussion of the square lattice model, in which the same methodological issues arise, in order to put the honeycomb lattice results in perspective.

In the square-lattice Hubbard model at half-filling, it is well known that the Mott transition is preempted by the onset of antiferromagnetic (AF) order. Nevertheless, the Mott transition may be investigated by quantum cluster methods if AF order is not allowed to set in. This is how it was observed in Ref. [9]. In that paper, the systems s4-4b and s4-8b (see Fig. 1) were treated with CDIA. Particle-hole symmetry left only one independent variational parameter in s4-4b: the cluster-bath hybridization parameter θ. In System s4-8b, a bath energy $\pm\varepsilon$ was also introduced, with opposite signs on the two bath orbitals linked to the same cluster site, so as to reflect particle-hole symmetry. A clear first-order Mott transition, with metallic, insulating, and unstable solutions, was found for the two systems, as shown in Fig. 2. The values of U_{c1} and U_{c2} are shown on these figures by vertical dashed lines.

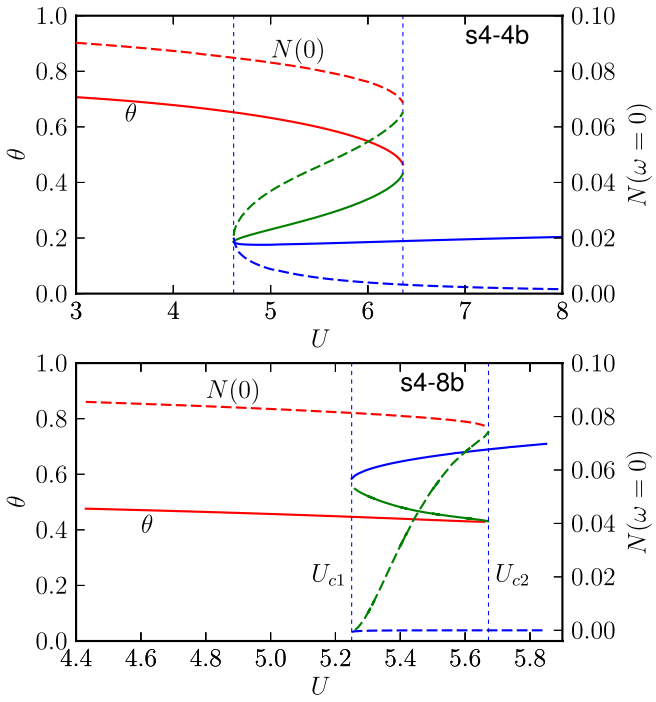

FIG. 2 (color online). Full line: Hybridization parameter θ as a function of U, obtained in CDIA for System s4-4b (top) and s4-8b (bottom). The three solutions (metallic, unstable, insulating) are shown. The vertical dashed lines indicate U_{c1} and U_{c2}. Dashed curve: density of states $N(0)$. Note the nonvanishing DoS in the "insulating" solution of s4-4b.

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 $A(\mathbf{k}, \omega)$ over wave vectors. We compute $N(\omega + i\eta)$ at $\omega = 0$ for a few values of the Lorenzian broadening η 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 ε 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 $\pm\varepsilon$.

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 μ, which would be the signature of a gap (the constraints on

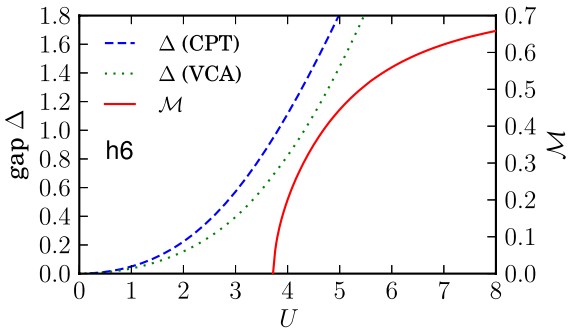

FIG. 3 (color online). Result of VCA calculations on the six-site cluster h6. The order parameter (red continuous curve) vanishes below $U = 3.75$. Blue dashed curve: spectral gap Δ computed without variational parameters (CPT). Green dotted curve: Δ computed from the VCA solution (t' used as variational parameter).

bath parameters stemming from particle-hole symmetry must then be released). This method does not require a Lorenzian broadening, as it involves integrals carried along the imaginary frequency axis. We have used both this method and the extrapolation method described above and find the same conclusions (see Supplemental Material [14]).

The lesson to carry from the square lattice is that a sound description of the Mott transition (hence of a putative spin liquid state) is to be found in a cluster system with minimally two bath orbitals per cluster site.

Let us now turn to the honeycomb lattice. Note that the antiferromagnetic order, on that lattice, is at zero wave vector. Thus the nesting condition is always satisfied and we expect 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 $\mathcal{M}$ 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

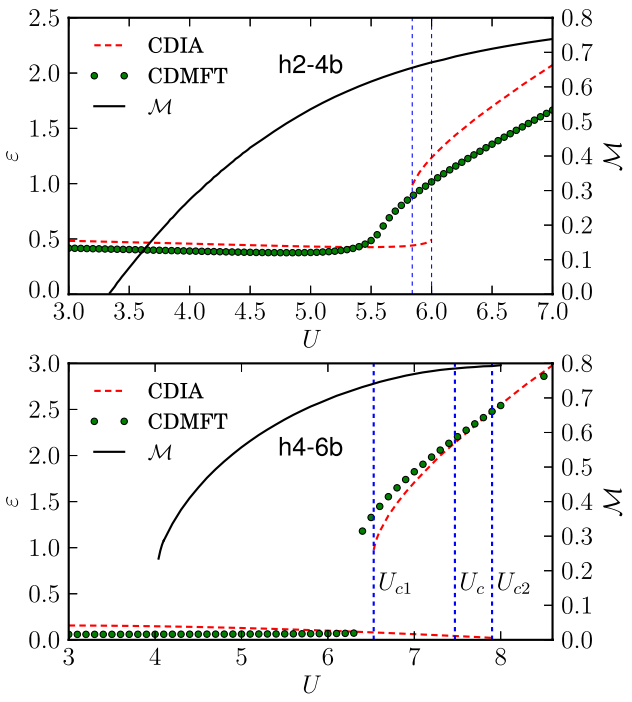

FIG. 4 (color online). Top: Solution of the h2-4b system. Bath energy ε obtained in CDIA (U_{c1} and U_{c2} are indicated by dotted vertical lines) and CDMFT. The staggered magnetization $\mathcal{M}$ obtained by CDIA is also shown. Bottom: Same, for System h4-6b. The value U_c where the metallic and insulating solutions have the same energy, is also indicated.

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 ($\pm \varepsilon$). In the nonmagnetic state, the two sites of the cluster (and the correponding bath sites) are related by left-right symmetry, and therefore only two bath parameters remain: one bath energy ε and one hybridization parameter θ. The CDMFT paramagnetic solution for ε is shown in the upper panel of Fig. 4. An upturn in the value of ε 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_{c2} and U_{c1}, and coexist in the range $[U_{c1}, U_{c2}]$. The first-order character of the solution is therefore clearly seen in CDIA.

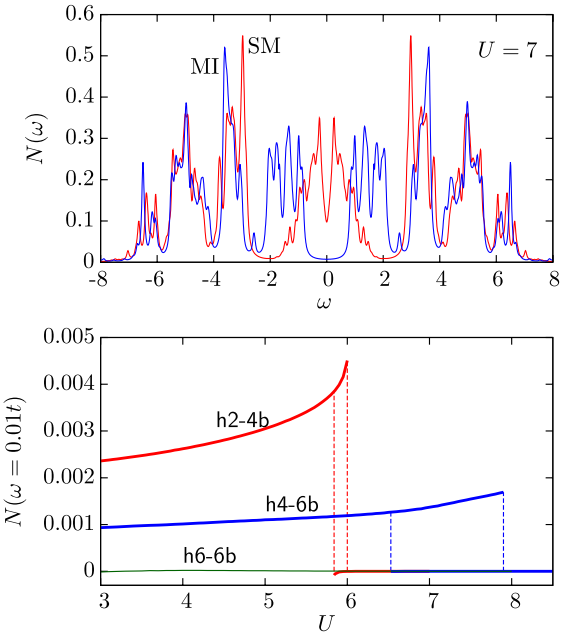

FIG. 5 (color online). Top panel: Density of states in the semimetallic (SM, red) and insulating (MI, blue) solutions obtained from System h4-6b at $U = 7$, with a Lorenzian broadening $\eta = 0.05t$. Bottom panel: Density of states $N(\omega = 0.01t + i\eta)$, extrapolated to $\eta \to 0$ for different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

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, $N(\omega + i\eta)$ is computed at $\omega = 0.01t$ instead of $\omega = 0$, since $N(0)$ is expected to vanish in the semimetallic phase.

Also shown in Fig. 4 is the antiferromagnetic order parameter obtained if the Weiss field M is added to the list of variational parameters; this constitutes in fact a mixture of CDIA and VCA, since the parameters at play within Potthoff's variational approach are both bath related (ε and θ) and cluster related (M). We find that, in this system, the critical interaction strength for the onset of magnetic order is $U \approx 3.4$, an even smaller value than in the six-site cluster VCA computation. Thus in this system the (continuous) magnetic transition preempts the Mott transition and no spin liquid occurs.

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, ε and θ, 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 $U \approx 6.35$, 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 Ω 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 h4-6b.
[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).