Variational study of triangular lattice spin-1/2 model with ring exchanges and spin liquid state in κ-$(ET)_2Cu_2(CN)_3$

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(Received 28 January 2005; published 5 July 2005)

We study triangular lattice spin-1/2 system with antiferromagnetic Heisenberg and ring exchanges using variational approach focusing on possible realization of spin-liquid states. Trial spin liquid wave functions are obtained by Gutzwiller projection of fermionic mean-field states and their energetics is compared against magnetically ordered trial states. We find that in a range of the ring exchange coupling upon destroying the antiferromagnetic order, the best such spin liquid state is essentially a Gutzwiller-projected Fermi sea state. We propose this spin liquid with a spinon Fermi surface as a candidate for the nonmagnetic insulating phase observed in the organic compound κ-$(ET)_2Cu_2(CN)_3$, and describe some experimental consequences of this proposal.

DOI: 10.1103/PhysRevB.72.045105 PACS number(s): 75.10.Jm, 75.50.Ee, 75.40.−s, 74.70.Kn

I. INTRODUCTION

This paper reports a variational study of spin-1/2 Heisenberg antiferromagnet with ring exchanges on a triangular lattice. One motivation for this study is the exact diagonalization work of LiMing *et al.*[1] and Misguich *et al.*[2] on this system proposing that it realizes spin liquid states. We are particularly interested in spin liquids that may occur near the Heisenberg antiferromagnetic state. Multiple-electron exchanges are believed to be important near quantum melting and metal-insulator transitions. The specific model considered here may also be relevant for the description of a tentative spin liquid state observed in the quasi-two-dimensional organic compound κ-$(ET)_2Cu_2(CN)_3$,[3] which is close to metal-insulator transition. Imada *et al.*[4] studied appropriate Hubbard model on the triangular lattice and found an insulating regime with no spin order. The ring exchange spin model can be viewed as derived from the Hubbard model by a projective transformation, which is appropriate in the presence of the charge gap.

The present work attempts to understand possible spin liquid states in the ring exchange model by examining candidate ground-state wave functions. This is complementary to the exact diagonalization studies, since knowing the character of a candidate wave function can give significant intuition.

The model Hamiltonian on the triangular lattice is, in the notation borrowed from Ref. 2,

$$\hat{H}_{\text{ring}} = J_2 \sum_{\bullet\!-\!\bullet} P_{12} + J_4 \sum_{\diamond} \left(P_{1234} + P_{1234}^{\dagger} \right) . \tag{1}$$

The two-spin exchanges are between all nearest neighbors and reduce simply to Heisénberg interactions $P_{12}=P_{12}^{\dagger}=2\boldsymbol{S}_1\cdot\boldsymbol{S}_2+\frac{1}{2}$. The four-spin "ring exchanges" are around all rhombi of the triangular lattice.

In the following, we consider only antiferromagnetic coupling $J_2>0$ and positive $J_4\geq 0$; for brevity, we set $J_2=1$. When $J_4=0$, the system is the familiar Heisenberg antiferromagnet on the triangular lattice and has a three-sublattice antiferromagnetic (AF) order. Exact diagonalization study of Ref. 1 proposes the phase diagram summarized in Fig. 1. The AF order is preserved for small $J_4\lesssim 0.07-0.1$, but is destroyed for larger J_4 and a spin gap opens up. However, in the regime $0.1\lesssim J_4\lesssim 0.25$ reported in Ref. 1, there are apparently many singlet excitations below the spin gap. Also, the spin gap starts to decrease for $J_4\gtrsim 0.175$.

In the exact diagonalization studies, it is hard to say which physical state is realized in the absence of clear signatures of some particular phase. The question of possible spin liquid states is taken up here by considering variational spin liquid wave functions on the triangular lattice. Specifically, we consider one family of such states obtained by Gutzwiller-projecting singlet fermionic mean-field states.[5,6] We determine the result of the competition with the AF ordered state by comparing against variational wave functions with long-range magnetic order.[7]

The result of the variational study is summarized in Fig. 2. For small $J_4\lesssim 0.14$, the AF state is stable compared with the tried spin liquid states. For larger J_4, we find spin liquid states that have lower variational energy than the magnetically ordered state. For example, we find that projected superconductor *Ansätze* perform well in the regime $0.14\lesssim J_4\lesssim 0.3$. More specifically, *Ansätze* with anisotropic extended s-wave, $d_{x^2-y^2}$, and $d_{x^2-y^2}+id_{xy}$ pairing patterns have very close optimal energies and much lower than the energy of the trial AF state. Unfortunately, we conclude that the present study is not sufficient to address the nature of the spin liquid in this regime, which we indicate with question marks in the figure. Our observation that the improvement in the trial energy is little sensitive to the specific pairing pattern may be an indication that the present restricted study cannot access the correct ground state in this regime.

A more robust conclusion from our study of such spin liquids is that the best *Ansätze* are close to the projected Fermi sea state and become more so for increasing J_4. Thus, for $J_4\gtrsim 0.3-0.35$ the variational Δ in our *Ansätze* reduces to essentially zero (below few percent of the hopping amplitude), and the ground state is essentially the projected Fermi sea.

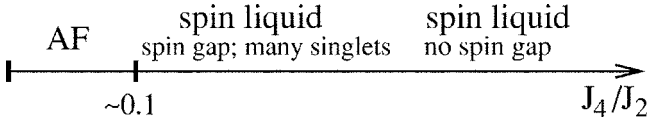

FIG. 1. Phase diagram of the model (1) from the exact diagonalization study of Refs. 1 and 2. The magnetic order is destroyed for $J_4 \gtrsim 0.07-0.1$; a spin gap is observed in the regime $0.1 \lesssim J_4 \lesssim 0.25$, but also many singlets below the spin gap. The spin gap is decreasing for $J_4 \gtrsim 0.175$.

The aptitude of the projected Fermi sea state can be intuitively understood as follows. We can view the ring exchange term with positive J_4 as arising from the electron hopping in the underlying Hubbard model (which we assume is in the insulating phase). Therefore, such ring exchange $J_4>0$ wants the fermions to be as "delocalized" as possible, and this "kinetic energy" is best satisfied in the simple hopping *Ansatz*. A more formal mean-field argument is given in Sec. III.

We now discuss the indications of this study for the possible spin liquid state in κ-$(ET)_2Cu_2(CN)_3$. This material is close to the metal-insulator transition, so the role of the electron kinetic energy is clearly important. Based on the experience with the ring exchange model, we therefore propose that the projected Fermi sea state is a good candidate ground state close to the metallic phase. We verify this more explicitly by considering a model with ring exchanges obtained by a projective transformation of the triangular lattice Hubbard model at order t^4/U^3. For the κ-$(ET)_2Cu_2(CN)_3$ compound we estimate $J_4/J_2 \simeq 0.3$. The results are summarized in Fig. 4. The work of Ref. 4 on the triangular lattice Hubbard model can be interpreted as an elaborate numerical study building up on free-fermion states, and hints some support to the proposed projected Fermi sea phase.

The proposed picture has many physical consequences. We have a Fermi surface of spinons, and therefore expect no spin gap and finite spin susceptibility down to zero temperature consistent with the experimental observations. An accurate treatment of the no-double-occupancy constraint and fluctuations requires that the spinons are coupled to a fluctuating U(1) gauge field. Such spinon-gauge field system has been studied extensively and is expected to exhibit some unusual behavior.[9–16] For example, one expects a singular contribution to the specific heat $C_{\rm sing} \sim T^{2/3}$ at low temperatures in two dimensions; the corresponding enhancement in "spin entropy" has concrete consequences for the phase boundaries.

The rest of the paper is organized as follows. In Sec. II we specify the variational states considered in this work. In Sec. III we seek qualitative understanding of the ring exchange energetics by considering a fermionic large-N treatment of the ring exchange Hamiltonian. In Sec. IV we consider the connection with the triangular lattice Hubbard model and possible application to κ-$(ET)_2Cu_2(CN)_3$. In particular, we describe experimental signatures of the proposed spinon Fermi surface-gauge system.

II. VARIATIONAL STATES AND ENERGETICS

In this section, we describe variational states used in the present work. Trial spin liquid states are constructed by

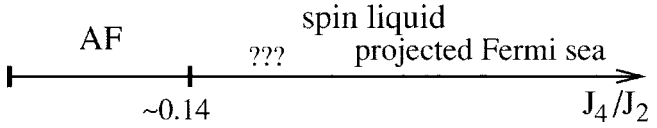

FIG. 2. Variational phase diagram for the Hamiltonian (1). The AF ordered variational state has the lowest energy for small J_4, but becomes unstable for $J_4 \gtrsim 0.14$ compared with the fermionic spin liquid states. One example of such spin liquid is the projected $d_{x^2-y^2}+id_{xy}$ superconductor *Ansatz*, with the optimal variational parameter $(\Delta/t)_{\rm var}=0.22$, 0.13, 0.05, 0.02 for $J_4=0.15$, 0.20, 0.25, 0.30, respectively. Some other *Ansätze* give very close optimal energy, and the situation is particularly not clear near the AF state. But for $J_4 \gtrsim 0.3$, our best *Ansätze* become essentially the projected Fermi sea state. We caution that for significantly larger J_4 states with more complicated magnetic orders—e.g., with four-sublattice order—may enter the energetics competition (Ref. 8), which is not considered here.

Gutzwiller projection of fermionic mean-field states.[5] We compare their energetics against AF ordered trial wave functions constructed using the approach of Huse and Elser.[7]

Spin liquid trial states. The starting point here is the fermionic mean field treatment of the Heisenberg model. A recent and very detailed description can be found in Ref. 5. The setup for constructing trial wave functions is as follows. Each spin operator is written in terms of two fermions $c_{r\uparrow}$ and $c_{r\downarrow}$, $\boldsymbol{S}_r = c_r^\dagger (\sigma/2) c_r$, with precisely one fermion per site. Heisenberg exchange interaction is written as a four-fermion interaction, which is then decoupled in the singlet channel. A convenient formulation of the mean field is to consider general spin rotation invariant trial Hamiltonian

$$H_{\rm trial} = -\sum_{rr'} \left[t_{rr'} c^\dagger_{r\sigma} c_{r'\sigma} + (\Delta_{rr'} c^\dagger_{r\uparrow} c^\dagger_{r'\downarrow} + \text{H . c . }) \right], \quad (2)$$

with $t_{r'r}=t^*_{rr'}$, $\Delta_{r'r}=\Delta_{rr'}$. For each such trial Hamiltonian we obtain the corresponding ground state. An SU(2) invariant formulation of the single occupancy constraint is that the isospin operator $\boldsymbol{T}_r \equiv \psi_r^\dagger (\tau/2) \psi_r$ is zero on each site; here $\psi_{r\uparrow}=c_{r\uparrow}$, $\psi_{r\downarrow}=c^\dagger_{r\downarrow}$, and τ^{1-3} are Pauli matrices. In the mean field, we require that this constraint is satisfied on average, which is achieved by tuning appropriate on-site terms. Going beyond the mean field, the physical spin wave function is obtained by projecting out double occupation of sites.

Many such trial states can be constructed, but there is also a gauge redundancy in this construction. Here, one is helped considerably by the recently available classification scheme of Wen[5,6] that allows one to construct all possible such fermionic mean-field states that lead to physically distinct spin liquids with specified lattice symmetries.

We numerically evaluate the expectation values of the two-spin and four-spin exchanges in such states using standard determinantal wave function techniques (so-called variational Monte Carlo).[17] We consider *Ansätze* with different sets of lattice symmetries, with and without time reversal, but primarily we focus on the nearest-neighbor *Ansätze* that respect upon projection the lattice translation symmetry. We then vary the parameters to optimize the trial energy.

AF ordered trial states. We want to compare the ring ex-

change energetics of the spin liquid states with the energetics of the antiferromagnetically ordered states. For this purpose, we use the family of variational states considered by Huse and Elser,[7] which capture well the Heisenberg model energetics. Our primary goal here is to see how the AF state is disfavored by the ring exchanges. Since we are comparing with rather different states and are looking for the energy level crossing, we do not need to know the ground-state energy very accurately, and the wave functions of Ref. 7 should be sufficient to get rough idea of the ring exchange energetics in the AF state. For details on these wave functions and numerical evaluations, the reader is referred to the original paper.

Variational results. We compared the trial energies of the AF ordered states and the fermionic spin-liquid states, and the result is summarized in Fig. 2. For small J_4, the ordered states have lower energy, but for $J_4 \gtrsim 0.14$ the spin liquid states win. The optimal spin liquid *Ansätze* have the following structure. The dominant part is the uniform triangular lattice hopping $t_{rr'}$, and for $J_4 \gtrsim 0.3-0.35$ we essentially find the projected Fermi sea state. In the intermediate regime $0.14 \lesssim J_4 \lesssim 0.3$, we find that the trial energy is improved upon adding $\Delta_{rr'}$ correlations into the mean-field wave function. Somewhat perplexingly, we find that the result is not very sensitive to the specific "pairing" pattern. Thus, optimized wave functions with extended anisotropic s-wave, $d_{x^2-y^2}$, and $d_{x^2-y^2}+id_{xy}$ pairing patterns have close energies. This may be an indication of an instability towards a state that cannot be captured in the context of the trial fermionic states. The situation is particularly inconclusive close to the AF phase, where several other trial states have competitive energies.

In summary, we find that ring exchanges disfavor the AF ordered state compared with the fermionic spin liquid states, but our study is not conclusive as to which spin liquid state is realized when the transition happens. Away from the transition, we suggest that the optimal spin liquid state is the projected Fermi sea state.

III. FERMIONIC LARGE N STUDY OF THE RING EXCHANGE ENERGETICS

In this section, we present a fermionic large N study of the ring exchange Hamiltonian. Here, natural "trial" states are pure hopping states, and this approach gives us some insight into their energetics. In particular, it shows how the ring exchanges favor the uniform hopping state, i.e., the projected Fermi sea state. The treatment below was suggested to the present author by Senthil. The mean-field analysis is also rather similar to an early work of Ioffe and Larkin.[18]

Consider the following generalization of the ring exchange Hamiltonian (1) to an SU(N) spin model

$$\hat{H}_{\mathrm{SU}(N)} = \frac{J}{N}\sum_{\langle 12\rangle}(c^\dagger_{1\alpha}c_{1\beta})(c^\dagger_{2\beta}c_{2\alpha}) + \frac{K}{N^3}\sum_P\left[(c^\dagger_{1\alpha}c_{1\beta})(c^\dagger_{2\beta}c_{2\gamma})\times(c^\dagger_{3\gamma}c_{3\delta})(c^\dagger_{4\delta}c_{4\alpha}) + \mathrm{H.c.}\right].$$

We use conventional fermionic representation with N fermion flavors; spin states on each site are viewed as states of $N/2$ fermions, i.e., we have occupancy constraint

$$c^\dagger_{r\alpha}c_{r\alpha} = N/2 \tag{3}$$

for each site r. In the above, summation over repeated flavor indices is implied. Our generalization of the exchange operators preserves the character of moving spins around a ring. For $N=2$, this Hamiltonian reduces precisely to the spin-1/2 Hamiltonian (1) with

$$J = 2J_2, \quad K = 8J_4. \tag{4}$$

A similar large N formulation was considered in a different context in Ref. 19. We also remark here that the general N formulation allows nontrivial exchanges involving three spins. This is unlike the $N=2$ case where such three-spin exchange reduces to a combination of two-spin exchanges. The three-spin exchanges can be easily included in the following analysis; to stay in line with the rest of the paper, we only consider the two-spin and four-spin exchanges.

We formulate the large N procedure in the spirit of the variational approach. Consider a single-particle "trial" Hamiltonian

$$\hat{H}_{\mathrm{trial}} = -\sum_{\langle rr'\rangle}(t_{rr'}c^\dagger_{r\alpha}c_{r'\alpha} + \mathrm{H.c.}) - \sum_r \mu_r c^\dagger_{r\alpha}c_{r\alpha}.$$

We find the ground state and use it as a trial wave function for the Hamiltonian $\hat{H}_{\mathrm{SU}(N)}$. In the mean field, the occupancy constraints are implemented on average by tuning the chemical potentials μ_r. The trial energy to leading order in $1/N$ is given by

$$\frac{E_{\mathrm{MF}}}{N} = -J\sum_{\langle 12\rangle}|\chi_{12}|^2 - K\sum_P(\chi_{12}\chi_{23}\chi_{34}\chi_{41} + \mathrm{c.c.}),$$

where $\chi^*_{rr'} \equiv \langle c^\dagger_r c_{r'}\rangle$ is the single-species expectation value.

We now have to minimize E_{MF} over the possible $t_{rr'}$ in the trial Hamiltonian. This leads to the following self-consistency conditions:

$$\Lambda^{-1}t_{rr'} = J\chi_{rr'} + \sum_{P=[1234]=[rr'34]} K\chi^*_{23}\chi^*_{34}\chi^*_{41}, \tag{5}$$

where the last sum is over all ring exchange plackets that contain the bond $\langle rr'\rangle$ as one of the consecutive bonds. Also, we have explicitly indicated the fact that the trial energy does not depend on the absolute scale in the trial Hamiltonian but only on the relative pattern of $t_{rr'}$.

We first make some general observations about this procedure. First of all, note that the self-consistency conditions imply that the optimal state can have nonzero $t_{rr'}$ only on the bonds that have nonzero $J_{rr'}$ or that appear in some ring exchange placket. For the triangular lattice model studied here, we then have to consider only nearest-neighbor $t_{rr'}$. Second, we see quite generally that the ring exchange contribution for a given placket has the form $-K|\chi|^4\cos(\Phi_P)$, where $|\chi|$ is the geometric mean of the absolute values of $\chi_{rr'}$ around the placket, while Φ_P is the "flux" of the corresponding phase factors. Thus, the positive ring exchange wants to smear the fermions over the lattice with no fluxes.

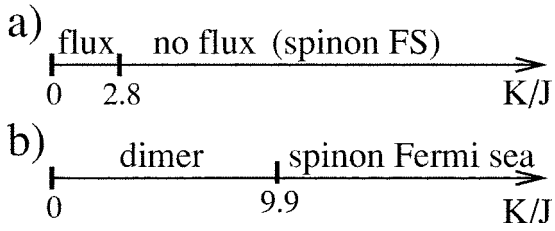

FIG. 3. Summary of the large N study of the Hamiltonian $\hat{H}_{\mathrm{SU}(N)}$. (a) Phase diagram from the mean-field energy optimization over translationally invariant states. (b) Full optimization.

To be more precise, let us consider several simple trial states. The uniform flux state has flux ϕ through each triangle. The expectation values $\chi_{rr'}=\langle c_{r'}^{\dagger}c_r\rangle$ have the same pattern of fluxes as the input $t_{rr'}$, and the trial energy per site is

$$E_\phi = -3J|\chi_\phi|^2 - 6K|\chi_\phi|^4\cos(2\phi), \tag{6}$$

since the flux through each rhombus is 2ϕ. Among such flux states, we find that for $K\lesssim 2.76J$ the best state has $\pi/2$ flux through each placket (this state has the largest $|\chi_\phi|$), while for $K\gtrsim 2.76J$ the best state has zero flux The numerical values of the energy per site in the two states can be obtained from

$$E_{\phi=\pi/2} = -0.120J + 0.0096K, \tag{7}$$

$$E_{\phi=0} = -0.081J - 0.0044K. \tag{8}$$

For large enough K/J the zero-flux state is stable against adding small flux ϕ because $|\chi_\phi|^2/|\chi_0|^2 \lessapprox 1+0.2\phi^2$.

We also considered so-called dimer states such that nonzero $t_{rr'}$ form nonoverlapping dimer covering of the lattice. These states break translational invariance, and any dimer covering produces such a state. It is well known that these states can have lower Heisenberg exchange energy in the large N limit. This is because the occupied bonds attain the maximal expectation value $|\chi_{rr'}|_{\max}$ and their contribution can be sufficient to produce the lowest total energy. The energy per site in any dimer state is

$$E_{\mathrm{dimer}} = -0.125J, \tag{9}$$

and is indeed the lowest energy for $K=0$. However, the dimer states gain no ring exchange energy, and for $K\gtrsim 9.9J$ the zero flux state becomes the lowest energy state. Finally, the so called box states have identical two-spin exchange energy with the dimer states but also nontrivial fluxes and therefore do not enter the competition for the ground state for $K>0$.

We performed full optimization over $t_{rr'}$ of the mean-field energy considering possible unit cells with up to four sites, and found that the above simple states are indeed sufficient to describe the ground state in the large N limit: The optimal state is one of the dimer states for $K\lesssim 9.9J$ and becomes the zero flux state for larger K. The complete study is summarized in Fig. 3.

To make better connection with the spin-1/2 system, we remark that we expect the dimer states to be energetically disfavored even for small K in the spin-1/2 case, e.g., compared with the flux states discussed above. This is because the Gutzwiller projection enhances local antiferromagnetic correlations more strongly in the translationally invariant mean-field states than in the dimerized states.[20] More quantitatively, the enhancement factor for the Heisenberg energy is roughly $g_J^{\mathrm{transl\ inv}}=4$ for the translationally invariant states, while it is only $g_J^{\mathrm{dimer}}=2$ for the dimer states. Furthermore, we expect even stronger enhancements in the ring exchange energy upon the projection. Taking all this into account, we expect the Fermi sea state to be favored for rather moderate J_4/J_2 in the spin-1/2 system.

To conclude the mean field discussion, our main message is that the positive ring exchange dislikes the fluxes and wants to make the system as uniform as possible. This is best realized in the projected Fermi sea state.

Going beyond the mean field, we obtain a theory of fermions coupled to a fluctuating gauge field $(a_0,\boldsymbol{a})$, where the temporal $a_0(r,\tau)$ enforce the local occupancy constraints while the spatial components represent the relevant fluctuations of $t_{rr'}(\tau)\approx|t|e^{ia_{rr'}(\tau)}$. The corresponding continuum theory ("relativistic electrodynamics in a metal") was studied in Refs. 9–16, and we will quote some results in the next section.

In the Appendix, we study long-distance properties of the Gutzwiller-projected wave function in some detail. As mentioned in the Appendix, this wave function may be not sufficient to capture the long wavelength behavior of the actual phase, since the projection treats only the a_0 fluctuations, but does not include the fluctuations of $a_{rr'}$, while the latter are crucial in the effective theory.[9–16] This is pointing a possible limitation of the projected wave function approach for the spinon-gauge system. We still expect that the variational study of the previous section gets the crude energetics correctly in the ring exchange model. This is also what we expect from the mean-field treatment, and leads us to propose the effective spinon-gauge theory. A finer numerical application likely requires more advanced techniques, perhaps in the spirit of Ref. 4 for the triangular Hubbard model. It would be interesting, for example, to look for the $2k_F$ signature[13] in the more elaborate work of Ref. 4, which may be a more accurate realization of the spinon-gauge ground state.

IV. APPLICATION TO POSSIBLE SPIN-LIQUID STATE IN κ-$(ET)_2Cu_2(CN)_3$

We now discuss possible spin liquid state in the organic compound κ-$(ET)_2Cu_2(CN)_3$, which is insulating and shows no magnetic order down to the lowest experimental temperatures. It is believed[3,4,21] that the conducting layer of this material is well described by a single-band triangular lattice Hubbard model at half filling with $t/U\simeq 1/8$ and only small hopping anisotropy of about 6%.

Unlike the square lattice case, for the half-filled triangular lattice we expect a metallic phase for large enough t/U. Reference 4 estimates the metal-insulator transition to occur at $(t/U)_{MI}\simeq 1/5$, so the κ-$(ET)_2Cu_2(CN)_3$ material is on the insulating side. Using an elaborate numerical technique, Ref. 4 finds a nonmagnetic insulator in this regime. We want to develop some picture of this state.

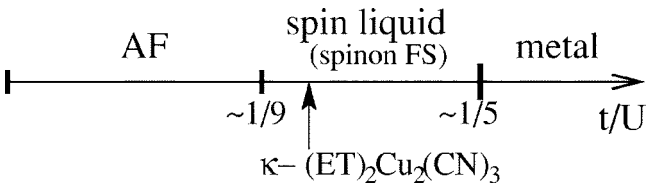

FIG. 4. Proposed phase diagram for the triangular lattice Hubbard model. The present study is based on the effective spin Hamiltonian (10) and applies only to the insulating regime expected for $t/U \lesssim 1/5$ from Ref. 4. Close to the metal-insulator transition, we propose the spin liquid state with spinon Fermi surface. For smaller $t/U \lesssim 1/9$, the best state is AF ordered. The κ-$(ET)_2Cu_2(CN)_3$ compound has $t/U \simeq 1/8$

The ideology we pursue here is that the insulating phase can be described by an effective spin model. Since the system is close to the metal-insulator transition, it is not enough to stop at two-spin exchange interactions. Starting with the Hubbard model, the effective Hamiltonian to order t^4/U^3 was obtained in Ref. 22. Specialized to the triangular lattice, the spin Hamiltonian reads

$$\hat{H}_{\text{eff}} = \hat{H}_{\text{ring}}[J_2, J_4] + \sum_{\langle\langle ij\rangle\rangle} J'' \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \sum_{\langle\langle\langle ij\rangle\rangle\rangle} J''' \boldsymbol{S}_i \cdot \boldsymbol{S}_j. \quad (10)$$

Here H_{ring} is the ring exchange Hamiltonian (1) with $J_2 = (1 - 32t^2/U^2)2t^2/U$, $J_4 = 20t^4/U^3$. The effective Hamiltonian has additional Heisenberg exchanges $J'' = -16t^4/U^3$ between second neighbors (separated by a distance $\sqrt{3}$) and $J''' = 4t^4/U^3$ between third neighbors (separation 2 lattice spacings). Our grouping of the terms in the effective Hamiltonian is intended to make it look as close as possible to the ring exchange model studied in the previous sections.

For the κ-$(ET)_2Cu_2(CN)_3$ compound, we estimate $J_4/J_2 \simeq 0.3$, which puts the ring exchange model into the proposed spinon Fermi sea regime. Further neighbor interactions not included in the J_2-J_4 model do not modify this result, even though J'' and J''' are roughly of the same magnitude as J_4. This stability is because the corresponding further neighbor spin correlations are small in the spin liquid regime.

To proceed more systematically, we repeat the variational study with the effective Hamiltonian (10). The resulting phase diagram is shown in Fig. 4 in terms of the Hubbard model parameter t/U. From this study, we propose that the insulating ground state is the antiferromagnet for $t/U \lesssim 1/9$ (this corresponds roughly to the ring exchange parameter $J_4/J_2 \approx 0.2$-0.25). For larger t/U, our best trial state is essentially the projected Fermi sea state, and the variational Δ (which can be used to improve the trial energy slightly) is small already at the transition from the AF state. In the same figure, we also indicate the metallic phase expected for $t/U \gtrsim 1/5$.

It should be emphasized that we do not treat either Hamiltonian (10) or (1) as more realistic or less realistic, particularly since we are dealing with the system near the metal-insulator transition. The above variational study with $\hat{H}_{\text{eff}}$ is presented primarily to illustrate that our results are not destabilized by making the Hamiltonian "more realistic." We expect that our main prediction for the spin-liquid state close to the metal-insulator transition is robust, since the proposed Gutzwiller-projected Fermi sea state is even more favored by including further effects of the electron kinetic energy. Also, the results of Ref. 4 give us some indication on the stability of the proposed state, since that study is building up on free-fermion states.

Physical properties in the spin liquid phase with spinon Fermi surface. The effective description of the proposed phase has spinon Fermi sea coupled to a dynamically generated gauge field. It has been argued[9–16] that this spinon-gauge system is described by a nontrivial fixed point and shows unusual behavior, which can be tested in experiments. Below, we list some thermodynamic properties of this Mott insulator. This phase is in some sense the closest one can get to the Fermi liquid while remaining a charge insulator, and shares some properties with the metal due to the presence of the spinon Fermi surface, but also has some "non-Fermi-liquid" properties.

Thus, spin susceptibility is expected to approach a constant as temperature T goes to zero:

$$\chi_{\text{spin}}(T \to 0) \sim \mu_B^2 \nu_0. \quad (11)$$

This is a consequence of having gapless spinon excitations over the entire Fermi surface and is in fact observed in κ-$(ET)_2Cu_2(CN)_3$.[23] Here, ν_0 is the density of states at the "Fermi surface" in the spinon band structure determined by the spinon "hopping amplitude" t_{spinon}. The latter is set by the Heisenberg exchange energy $t_{\text{spinon}} \sim J$ and is different from the bare electron hopping amplitude t_{el} (remember that $J \sim t_{\text{el}}^2/U$). For the triangular lattice at half-filling, we have $\nu_0 = 0.28/t_{\text{spinon}}$ per triangular lattice site and including spin. Reference 3 reports $\chi = 2.9 \times 10^{-4}$ emu/mol at low temperatures, from which we estimate $t_{\text{spinon}} \approx 350$ K. This compares favorably with the reported magnitude of the Heisenberg exchange $J = 250$ K. Furthermore, Ref. 24 observes that ^{13}C nuclear spin relaxation rate $1/(T_1 T)$ approaches a constant at low temperature, which is what one expects for the spinon Fermi surface.

Specific heat, on the other hand, is expected to show non-Fermi-liquid behavior

$$C \sim k_B \nu_0 t_{\text{spinon}}^{1/3} (k_B T)^{2/3}. \quad (12)$$

This is written to contrast with the Fermi liquid $\sim T$ behavior, and means that the spin entropy in this charge insulator is in fact larger than in the metallic state at low temperature. This is very different from the antiferromagnet or gapped spin liquid insulators which have low spin entropy. In particular, the finite temperature first-order transition line between the proposed spin liquid and the metallic state is expected to bend towards the metallic state with increasing temperature[25,26]

$$p_{MI}(T) - p_{MI}(0) \sim T^{5/3}. \quad (13)$$

In the last formula, p is an applied pressure which drives the insulator to metal transition.[3,27] This tendency is actually observed in the κ-$(ET)_2Cu_2(CN)_3$ material.[28]

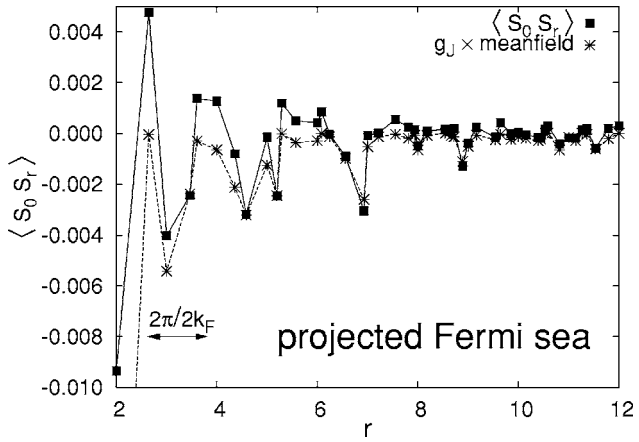

FIG. 5. Spin correlation in the projected Fermi sea state. Measurements are done on a 24×24 triangular lattice. The mean-field wave function is constructed for periodic boundary conditions and excluding the zero momentum single-particle state in order to avoid Fermi surface points while satisfying the lattice rotation symmetry for the finite system (this does not affect the long-distance properties of the wave function which is our focus here). Note the oscillating character of the correlation [with the period $\approx 2\pi/(2k_F)$, $k_F \approx 2.69$]. Also note that the renormalized mean field roughly reproduces the overall magnitude of the correlations.

V. CONCLUSIONS

In summary, we considered the spin-1/2 ring exchange model on the triangular lattice from the variational perspective and identified the instability of the antiferromagnetically ordered state towards spin liquid state in the regime of moderate ring exchange couplings. Our best trial states become the Gutzwiller-projected Fermi sea state for larger J_4. Despite the limitations of the variational approach, it is hoped that the present work may give complimentary information and useful guidance for understanding the exact diagonalization results.

We also studied the effective spin Hamiltonian appropriate for describing charge insulator states of the triangular lattice Hubbard model. The effective Hamiltonian includes Heisenberg exchanges as well as ring exchanges, and so is close to the considered ring exchange model. The study is motivated by the tentative spin liquid state in the κ-$(ET)_2Cu_2(CN)_3$ compound, which is modeled by the triangular lattice Hubbard model in the vicinity of the metal-insulator transition. We find that upon including the ring exchanges but well in the insulating regime, the antiferromagnet gives way to the spin liquid state which is essentially the projected Fermi sea state. In view of this finding, we propose that the effective description of the nonmagnetic insulator phase has Fermi sea of spinons coupled to the dynamically generated gauge field. This spin liquid phase features a number of unusual properties which can be looked for in experiments. It would be very exciting if this remarkable state is indeed realized in the κ-$(ET)_2Cu_2(CN)_3$ material.

Note added in proof. A recent preprint by Lee and Lee[29] studies half-filled triangular lattice Hubbard model using slave-rotor representation, and suggests the state with spinon Fermi surface as a candidate for the spin liquid observed in κ-$(ET)_2Cu_2(CN)_3$, similar to the present work. Reference 29 also discusses further experimental consequences for the proposed state, and in particular predicts a highly unusual temperature dependence of the thermal conductivity.[30]

ACKNOWLEDGMENTS

The author has benefited from many useful discussions with M. P. A. Fisher, V. Galitski, P. Nikolic, and A. Vishwanath, and is especially grateful to T. Senthil for motivating this problem and sharing many insights throughout the course of the study. This work was started at MIT and was supported by NSF grants Nos. DMR-0213282 and DMR-0201069. The work at KITP was supported through NSF grant No. PHY-9907949.

APPENDIX: PROPERTIES OF THE PROJECTED FERMI SEA WAVE FUNCTION

We describe some properties of the projected Fermi sea state. Figure 5 shows spin correlations in the projected wave function and also in the free fermion state before the projection. In the free fermion state, the spin correlation behaves as $-\cos^2(k_F r - 3\pi/4)/r^3$ at large distances, which oscillates with the wave vector $2k_F$ while always staying negative. To facilitate the comparison, Fig. 5 shows the mean-field correlations in the specific finite system (the finite size effects are fairly large because of the gaplessness over the Fermi surface). We observe that the effect of the projection is not strong: For the range studied, the mean-field result multiplied by the Gutzwiller enhancement factor $g_J=4$ gives a reasonable approximation for the actual correlation.[20] After the projection, the correlation function now swings to positive values as well, but the overall magnitude is roughly captured by the simple renormalization factor.

We also studied spin chirality correlations (not shown), and found that these are very small beyond few lattice spacings. The effective theory of the proposed phase has Fermi sea of spinons coupled to a dynamically generated gauge field.[9–15] The measured spin correlations in the projected wave function represent some puzzle in this respect: Ref. 13 predicts that the spin structure factor is singularly enhanced near $2k_F$ in the spinon-gauge system. We find that in the projected wave function the structure factor remains finite throughout the Brillouin zone and that the overall rate of decay of spin correlations is roughly the same as in the free fermion state. One possible source of this difference is that the projected wave function has fixed $t_{rr'}$ and therefore does not include the fluctuations of the spatial components of the gauge field; only the temporal component is "included" by the projection. This is a limitation of the projected wave function approach for the spinon-gauge system.

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