1 Mobile holes in frustrated quantum magnets and itinerant fermions on frustrated geometries

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1.1 Introduction

As discussed in several chapters of this volume, frustration leads to unconventional (insulating) ground states. On the other hand doped holes are known to have profound effects in Mott insulators. Therefore doped frustrated systems offer the prospect of novel phases with some of the most fascinating, challenging and exotic behaviour. In addition, at commensurate electron fillings and in the presence of strong (screened) Coulomb repulsion, geometrical frustration can also manifests itself as an extensive degeneracy of the classical ground-state manifold providing profound similarities with the field of quantum frustrated magnetism.

Magnetic frustration in quantum spin systems leads frequently to the formation of spin singlets (dimers). Generically, systems of fluctuating quantum dimers can often order, breaking lattice symmetries to give rise to Valence Bond Crystals (VBCs) [1], but under other circumstances they may remain in a quite unconventional quantum disordered state, the spin liquid, which breaks neither spin nor lattice symmetries. Anderson’s original $d$-wave Resonating Valence Bond (RVB) state [2] is a paradigm for the spin liquid (in fact, for a particular type of gapless spin liquid, while the short-range RVB state composed of only nearest-neighbor dimers is gapped spin liquid). In a number of cases, frustrated spin systems and/or dimer systems can be doped, for example by chemical substitution in a Mott insulator. When both spin and charge degrees of freedom are present, the role of frustration becomes unclear, and to date remains only poorly explored. It is, however, clear that new and exotic phenomena emerge upon doping, including heavy-fermion behavior, spin-charge separation or quasiparticle fractionalization, unconventional superconductivity, stripe formation, bond and/or charge ordering, and many
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Such fundamental issues have motivated an increasing number of recent investigations, as well as the continuing search for new, doped materials. In this chapter, we describe some selected topics which illustrate the richness and diversity of the field of doped, frustrated magnets. The first example concerns the dynamics of a small number of doped holes in the two-dimensional (2D) kagome and checkerboard Heisenberg quantum antiferromagnets. Without doping, the kagome Heisenberg antiferromagnet is believed to be a serious candidate for spin-liquid behavior, while the checkerboard lattice is the 2D analog of the well-known and highly frustrated 3D pyrochlore structure common in real materials. With doping, issues such as particle fractionalization and pairing can be addressed. In a second example, we move to the topic of Quantum Dimer Models (QDMs), similar to those proposed by Rokhsar and Kivelson in the context of the pseudo-gap phase of high-temperature superconductors. Two classes of (weakly) doped QDMs will be discussed, which differ in the assumed statistics, bosonic or fermionic, of the bare holes. We proceed further by considering strongly correlated electrons on frustrated triangular lattices, and discuss the physics of an unconventional, reentrant metal-insulator transition. As our final example, we consider correlated fermions moving on frustrated lattices at special, commensurate densities for which exotic but once again insulating ground states (GSs) are stabilized. For systems with strong interactions (Mott insulators), we show briefly how their behavior is analogous to the physics of (undoped) QDMs.

1.2 Doping holes in frustrated quantum magnets

1.2.1 The holon-spinon deconfinement scenario

We begin our discussion of the phenomena associated with doped holes in frustrated magnets by considering the most popular paradigm for a nonmagnetic quantum ground state, which is a dimer-based system. If we assume that a single hole is “injected,” then the removal of the electron results in the breaking of one of the dimers, leaving behind an empty site (holon) and a free spin (spinon) on the same bond.

If the dimers can change their positions by quantum fluctuations, the holon and the spinon can move on the lattice, across the diagonals of the plaquettes in a square-lattice system, by exchanging with the dimers. A typical configuration is shown in Fig. 1.1. Optimization of their kinetic energies would require the holon and spin to delocalize, and thus to become separated. In a (gapped) spin liquid, realized if the system can fluctuate through all possible dimer coverings, a complete separation, known as “deconfinement,” is possible. In this situation, the Landau quasiparticle (QP) breaks apart into separate species, and an experimental technique which probes the hole Green function, such as Angle-Resolved Photoemission Spectroscopy (ARPES), would then show a broad maximum in place of the sharp QP peak. However, if dimer VBC order...
is present, i.e. only one specific (type of) covering lies lowest in energy, one expects an effective string potential that binds the holon and spinon: indeed, if the dimers had no internal structure, an attempt to separate these two “particles” would lead to a continuous and linear increase of their energy. In reality, this increase is bounded by the spin gap (the energy to break up a dimer), beyond which which pairs of spinons would be generated spontaneously along the string.

1.2.2 Single hole doped in frustrated Mott insulators

Frustrated magnets are good candidates for the observation of spin-charge separation upon doping. The checkerboard and kagome lattices, shown in Figs. 1.2(a) and (c), are good examples of the types of frustrated lattice on which such a phenomenon may be expected. They are composed, respectively, of strongly frustrated tetrahedra and triangles, assembled in a 2D, corner-sharing structure. While the AF Heisenberg Hamiltonian for $S = 1/2$ spins (in this chapter we consider only systems of $S = 1/2$ spins) on the checkerboard lattice is now thought to be a fully gapped system exhibiting VBC order (plaquette phase) [3–6], by contrast no sign of ordering has been found in the undoped kagome antiferromagnet [7, 8], which possesses an exponentially large number of singlet states within the (finite-size) spin gap [8, 9]. It is these unconventional, low-lying excitations which open the door to new and surprising phenomena upon hole doping.

We perform exact diagonalization (ED) calculations based on the standard $t$-$J$ model Hamiltonian,

$$H_{t-J} = -t \sum_{\langle i,j \rangle, \sigma} \mathcal{P} \left( c_{i,\sigma}^{\dagger} c_{j,\sigma} + \text{h.c.} \right) \mathcal{P} + J \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_i n_j), \quad (1.1)$$

where on both lattices all bonds have the same couplings $t$, describing the kinetic energy of the hopping quasiparticles, and $J$, which is the superexchange
Fig. 1.2. (a) Checkerboard lattice. The two degenerate VBC GSs of the nearestneighbor Heisenberg model (half-filling) are represented schematically in blue and red. This figure also serves as a representation of the RSPC GSs of the quarter-filled, large-\( \sqrt{2} \times \sqrt{2} \) super-cell reflection. The path from the zone center (\( \Gamma \)) to the \( \mathbf{k} = (\pi, \pi) \) point (\( M \)) is shown as a dashed line. (b) Full Brillouin Zone (BZ) of the checkerboard lattice. The dotted line corresponds to the reduced BZ associated with the \( \sqrt{2} \times \sqrt{2} \) super-cell reflection. (c) Kagome lattice. (d) As in (b) but for the case of the kagome lattice.

interaction between the spins. This model is believed to be a reliable description of the low-energy physics of weakly doped Mott-Hubbard insulators with large optical excitation gaps. Here and hereafter, we assume the value \( J = 0.4 \) (in units where \(|t|\) is set to 1), which is the general order of the physical value in a number of real materials. The hole spectral functions are defined in the standard way as

\[
A(k, \omega) = -\frac{1}{\pi} \text{Im} \left[ \langle \Psi_0 | c_{k,\sigma}^\dagger \omega + E_0 + i\eta - H \mid \Psi_0 \rangle \right],
\]

and calculated by Lanczos ED, supplemented with a continued-fraction technique, on finite clusters with periodic boundary conditions to take advantage of the lattice translation symmetry. The reader is referred to the chapter of A. L"auchli in this volume for a detailed discussion of numerical methods. Here we focus on the case of a single dynamic hole, as studied in Ref. [10]. Because of the absence of particle-hole symmetry in frustrated lattices, it is necessary to distinguish between the cases \( t > 0 \) and \( t < 0 \). Note that for \( t < 0 \), frustration can also appear in the hole motion: in the example of a particle with the
tight-binding dispersion on an isolated triangle, the kinetic-energy gain is $|t|$, a factor of two smaller than for $t > 0$.

**Fig. 1.3.** Single-hole spectral functions obtained on a 32-site checkerboard cluster ($\sqrt{32} \times \sqrt{32}$, tilted at 45° to the axes of Fig. 1.2) along the line $\Gamma M$. Left panel: $t = +1$; right panel: $t = -1$. In both cases, $J/|t| = 0.4$. When a quasiparticle peak is present, the corresponding weight is indicated. Magnification factors applied in some cases are as indicated. From Ref. [10].

Typical results obtained for a 32-site cluster on the checkerboard lattice are shown in Fig. 1.3 for the line $\Gamma M$ line in the Brillouin Zone (BZ) [Fig. 1.2(b)]. At all points, most of the spectral weight is found to be incoherent, distributed over a range of 7-9$|t|$. However, a small QP peak is visible, particularly for momenta close to the $M$ point. The region close to the $\Gamma$ point has only a very small QP peak, or possibly none at all, and the shape of the spectral function at $\Gamma$ itself is very special, probably because of its higher point-group symmetry.

The analogous spectral functions of the kagome lattice, shown in Fig. 1.4, show definite exotic behavior; they are very broad for all momenta (widths approximately 6-8$|t|$) and, in contrast to the checkerboard lattice, show no visible QP peaks, either for $t > 0$ (left panel) or for $t < 0$ (right panel). We stress that the broad appearance of these spectra has no connection to the value of $\eta$ used in the calculation, but is an intrinsic feature of the spectral function, as can be deduced from the large number of poles carrying spectral weight (circles in Fig. 1.4). These spectral-function data support very strongly a spin-charge-separation scenario for the kagome lattice. Indeed, this spectacular phenomenon can be observed directly in the spin-density profile in the vicinity of the hole: a repulsion between the net $S = 1/2$ moment and the
mobile hole is clearly visible, providing further support to the deconfinement scenario described above in the context of dimer-based systems.

Fig. 1.4. Single-hole spectral functions (black lines) along the line $\Gamma \leftrightarrow M$, computed on a 27-site kagome cluster for $t = +1$ (left panels) and $t = -1$ (right panels). Contributions of both singlet and triplet final states are included (see text). The red circles denote pole locations and their residues. From Ref. [10].

This investigation provides the first example of the observation of spin-charge separation in a 2D microscopic model. It establishes that the spin-liquid nature of the undoped ground state is crucial for such behavior. Indeed, in the checkerboard lattice, whose ground state exhibits a VBC structure, a weak holon-spinon confinement manifests itself as QP peaks in the spectral function for some momenta.

1.2.3 Hole pairing and superconductivity

Whether doped holes could pair and lead to unconventional superconducting behavior is another of the fundamental issues raised recently by the new prospect of doping frustrated antiferromagnets. Indeed, superconductivity in the spinel oxide LiTi$_2$O$_4$ [11], in the recently synthesized 5$d$ transition-metal pyrochlores [12], and in a layered triangular CoO compound [13] suggests that geometrical frustration, which could be magnetic and/or kinetic, might play a key role in the mechanism of unconventional superconductivity (as discussed in the chapter of Z. Hiroi and M. Ogata).

Cluster calculations (Fig. 1.5) were used to discover the occurrence of pairing in the doped checkerboard Heisenberg antiferromagnet described above [14]. It was shown that pairing, in several orbital channels including $s$- and $d$-wave, appears at arbitrarily small $J/t$ for the particular sign of the
hopping amplitude which leads to frustration in the motion of a single hole. In
fact, hole delocalization (i.e. a gain in kinetic energy) plays a key role in this
new mechanism of unconventional pairing, as also in some of the inter-layer
tunneling mechanisms proposed by Anderson \cite{15}. From these numerical data,
a simple scenario might be proposed for $t > 0$: despite its suppressed coherent
motion, a single hole retains a strong incoherent motion, and thus can act to
“melt” the plaquette VBC in its vicinity. This region, which may be somewhat
extended in space, becomes more favorable for a second hole to gain
kinetic energy, leading to correlated (or assisted) hopping. It is interesting
to emphasize here the similarities with tight-binding studies of frustrated lattices,
which show both localized single-particle states and interaction-induced,
delocalized, two-particle bound states \cite{16}.

In connection with cobaltates, superconductivity has also been investi-
gated in the $t$-$J$ model on the triangular lattice, in particular using RVB
variational Ans"atze (presented in the chapter of Z. Hiroi and M. Ogata). In
these studies, $d_{x^2-y^2} + id_{xy}$-wave superconductivity is found to be stable near
half-filling. The relationship between this phase and the three-sublattice, 120-
degree AF long-range order occurring at half-filling remains at present unclear
and in need of further investigation.
1.3 Doped Quantum Dimer Model

1.3.1 Origin of the Quantum Dimer Model

The conclusions obtained in Sec. 1.2.2 notwithstanding, both magnetic frustration and the introduction of fermionic variables (holes) lead, together or independently, to severe limits on the available numerical approaches. For example, quantum Monte Carlo algorithms, known to be very efficient for simple quantum spin systems, suffer from the infamous “minus-sign problem” (introduced in the chapter of A. Läuchli) and cannot be used at the required low temperatures. Practically, zero-temperature ED (by the Lanczos algorithm) and variational approaches are the only controlled methods practicable for systems such as the $t$-$J$ model on frustrated lattices. However, one alternative route to the investigation of microscopic models of the $t$-$J$ and Hubbard types is to construct effective models which would allow the use of more efficient methods, or calculations on larger clusters, while retaining the essential low-energy physics.

When the effect of magnetic frustration is such that dimer degrees of freedom are relevant, one may consider the quantum hard-core dimer gas on a two-dimensional lattice. We illustrate this type of model by considering a square lattice, on which it is defined by the Hamiltonian

$$H_{\text{QDM}} = V \sum_c N_c |c\rangle \langle c| - J \sum_{\langle c, c' \rangle} |c\rangle \langle c'\rangle,$$

where the sum over the index $c$ refers to all nearest-neighbor dimer coverings, $N_c$ is the number of “exchangeable” plaquettes, and the sum $\langle c', c \rangle$ is over

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Fig. 1.6. (a) Dimer-exchange process and (b) holon hopping process in the QDM.
all pairs of configurations \(|c\rangle\) and \(|c'\rangle\) that differ by a single dimer-exchange process of the type illustrated in Fig. 1.6(a). In a manner similar to the square lattice, on the triangular lattice the exchange of parallel dimer pairs can be performed on the three different types of two-triangle rhombi. This model was introduced originally by Rokhsar and Kivelson [17] in the context of the RVB theory of cuprate superconductors. The connection to the original spin formulation is, however, not completely clear: among other truncations of the spin degrees of freedom, the QDM of Eq. (1.3) deals by construction with orthogonal dimer coverings, which is not the case for the SU(2) dimer basis relevant in frustrated Heisenberg antiferromagnets. In spite of these subtleties, QDMs are expected to capture the essential physics of systems with singlet ground states, one primary reason for this being that they do possess the extreme ground-state degeneracy of the basis manifold. More details concerning these issues may be found in the chapter of R. Moessner in this volume.

It is easy to introduce doping in the QDM. Holes may be injected only in pairs (i.e. by removing dimers). However, doped holes can then move independently by hopping between nearest-neighbor (on the triangular lattice) or diagonal next-nearest-neighbor (on the square lattice) sites [17–19]. The full Hamiltonian for a doped QDM is

\[
H = H_{QDM} - t \sum_{(c,c')} |c'\rangle\langle c|,
\]

where the \((c', c)\) sum involves all pairs of configurations \(|c\rangle\) and \(|c'\rangle\), containing a fixed number \(N_h\) of vacant sites (holes), that differ by a single hole hopping along a plaquette diagonal as illustrated in Fig. 1.6(b). In this formulation, bare holons, by which is meant the moving vacancies, have Bose statistics. Note that, in contrast to the triangular lattice, holes on the square lattice are constrained to remain only on one of the two sublattices.

**Fig. 1.7.** Schematic phase diagrams of the undoped QDM for both the triangular (top) and square (bottom) lattices. Only the triangular lattice is believed to exhibit a liquid (RVB) phase. Evidence for the mixed phase on the square lattice has been reported in Ref. [23].
1.3.2 Phase diagrams at zero doping

Somewhat remarkably, the square- and triangular-lattice QDMs have quite different phase diagrams in the undoped case. First, exactly at \( V = |J| \), which is known as the “RK point,” the GS takes the form of an equal superposition of all dimer coverings, and exhibits algebraic dimer correlations on the square lattice but short-ranged (exponentially decaying) correlations on the triangular lattice. Ordered VBC states appear on the square lattice immediately away from the RK point, whereas a gapped RVB liquid \([20]\) is present over a finite region in \( V/J \) on the triangular lattice. This RVB phase of the triangular lattice has also been shown to exhibit topological order \([20, 21]\), whose importance for frustrated systems is discussed in the chapter of G. Misguich, and for quantum information in the chapter of J. van den Brink, Z. Nussinov and A. M. Oleś. Comparative schematic phase diagrams for the two lattices are depicted in Fig. 1.7. The square lattice shows a rich variety of VBC phases \([22]\) with, in particular, a novel mixed phase \([23]\) which interpolates between the columnar and the plaquette phases (the blue squares in Fig. 1.7 correspond to plaquettes on which vertical and horizontal dimer pairs resonate).

1.3.3 Connection to the XXZ magnet on the checkerboard lattice

We have already explained the extent to which QDMs provide a natural framework to describe the dynamics of SU(2) singlets in frustrated but isotropic quantum antiferromagnets. We also mention briefly another case in which the QDM emerges as the model for the low-energy physics, that of the strongly anisotropic Heisenberg magnet (in the Ising limit) on the checkerboard lattice and in the presence of a magnetic field.

We begin with no magnetic field and only an Ising coupling, \( J_z S_i^z S_j^z \), on the bonds of the checkerboard lattice: in this case, the (classical) ground state is highly degenerate and can be fully represented by the “loop coverings” illustrated in Fig. 1.8(a). Here, an up- (down-)spin is represented by the presence (absence) of a dimer, or boson, on the bonds of an effective square lattice whose sites are in fact the centers of the “tetrahedra” (the squares including diagonal bonds). The constrained nature of the classical GS is of the “ice-rule” type: the lowest Ising energy is obtained when there are precisely two bosons on every tetrahedron. Second-order processes in the exchange coupling \( J_{xy} \) lead to the dynamics of a six-vertex model \([24]\) or a quantum loop model \([25]\).

By applying a magnetic field, the density of dimers (bosons) can be altered systematically. When an average of one dimer per tetrahedron is reached, again the ground-state manifold obeys an ice-rule constraint in the large-\( J_z \) limit, where all states with precisely one dimer on every tetrahedron, as shown in Fig. 1.8(b), are ground states. Second-order processes in \( J_{xy} \) now lead to a QDM on the effective square lattice with \( V = 0 \) and \( J = J_{xy}^2/J_z \) in Eq. 1.3.
1.3.4 Bosonic doped Quantum Dimer Model

We turn now to the doped QDM and concentrate first on the case \( J > 0 \) in Eq. 1.4. In the mapping from SU(2) dimers, this sign of \( J \) is expected for a bosonic representation of the singlet bonds. The “monomers” (holes) of the doped QDM discussed here could then be interpreted physically as entities such as unbound spinons.
For $J > 0$ and $t > 0$, the off-diagonal matrix elements of the Hamiltonian (1.4) are all non-positive, so that (from the Perron-Frobenius theorem) its GS has no node. Consequently, Green-function Monte Carlo (GFMC) techniques can be applied, particularly in the vicinity of the RK point and for small $t/J$ ratios (i.e., when the exact RK GS is still a good guiding wave function), and the phase diagrams shown in Fig. 1.9(a,b) can be extracted by appropriate finite-size scaling. For larger values of $t/J$, such calculations can be complemented by ED on smaller clusters [26]. The phase-separation (PS) region consists of phase coexistence between an undoped VBC and a superfluid, the latter becoming stable as a unique component above a critical doping. It is notable that this superfluid exhibits flux quantization in units of $\hbar/2e$, in qualitative agreement with gauge theories of high-temperature superconductors [27] and recent, related $Z_2$ gauge theories [28].

Fig. 1.10. Schematic phase diagrams of the non-Frobenius ($J < 0$) doped QDM as a function of dopant concentration $x$ and $V/|J|$ (a) or $|J/t|$ (b), estimated by ED calculations [26].

1.3.5 Non-Frobenius doped Quantum Dimer Model on the square lattice

Turning to the case of Eq. 1.4 with $J < 0$, a quite different type of behavior is expected. The “non-Frobenius” nature of the Hamiltonian (which prohibits the use of GFMC) reflects the original “Fermi sign” of the strongly correlated electrons. Indeed, if one interprets the dimers as SU(2) singlets, a dimer creation operator on the bond $ij$ can be written in the fermionic representation [29,30] as $d_{ij}^\dagger = (f_{i1}^\dagger f_{j1}^\dagger + f_{i1}^\dagger f_{j1}^\dagger)/\sqrt{2}$. In this basis, it can be verified that the effective dimer-exchange process generated by the underlying Heisenberg interaction within a plaquette occurs for the sign $J < 0$ [29]. In addition,
the electron-destruction operator takes the form \( c_{i\sigma} = f_{i\sigma} b_i^\dagger \), where the holon (hole or monomer) creation operator \( b_i^\dagger \) is bosonic.

The phase diagram of the non-Frobenius doped QDM obtained by ED [26] is both exotic and rich, as shown in Fig. 1.10. First, bare holons can be seen binding to topological defects (namely vortices, also known as “visons”), producing fermionic composite particles; alternatively stated, the hole becomes a fermion. Secondly, in contrast to the bosonic case, no PS is seen in the immediate vicinity of the (VBC) Mott insulator. Instead, a \( d \)-wave pairing is expected, opening the possibility of unconventional superconductivity. Finally, at large kinetic energies, holons and vortices unbind, bosonic holes Bose-condense, and a superfluid phase is stabilized, presumably of the same type (with \( 2e \) charge quanta) as that obtained for \( J > 0 \).

1.4 Mott transition on the triangular lattice

1.4.1 Frustration in itinerant electron systems

In the first half of this chapter, we have discussed the dynamics of holes doped into magnetic insulators with geometrical frustration. Another important class of phenomena is driven by frustration in metallic systems. Several strongly correlated metallic systems, such as LiV\(_2\)O\(_4\) [31] and (Y,Sc)Mn\(_2\) [32], show unusually large entropies at temperatures much lower than their bare energy scales (band width and Coulomb repulsion), and this is thought to be related to the geometrical frustration inherent in their lattice structure [33, 34]. In strongly correlated electron systems, double occupancy of a site is suppressed by the large, on-site Coulomb repulsion, and the probability of single occupancy increases. This tends to enhance the formation of a local magnetic moment at each site, which interacts with neighboring moments, and frustrated configurations may be adopted depending on the lattice geometry. The central issues for frustrated metals are the effects of exotic magnetic fluctuations on quasiparticle coherence and novel magnetic long-range order or characteristic correlations in itinerant systems.

1.4.2 Mott transition in organic compounds with triangular geometry

One of the well-studied problems in the physics of frustrated metals is the Mott metal-insulator transition on a triangular lattice [35–37]. When the Coulomb repulsion is much larger than the band width \( (U \gg W) \), the half-filled system is described effectively by the Heisenberg spin model in the sector of energies below the Mott-Hubbard charge gap. In this case, the well-known 120° structure appears in the ground-state spin configuration. A more exotic situation can be expected when the Coulomb repulsion is comparable with the band width \( (U \sim W) \), when ring-exchange processes involving multiple sites
become important. These processes, which are discussed in the chapter by G. Misguich, open the possibility of stabilizing exotic states.

Experiments on organic compounds with a triangular lattice structure [38–40] have stimulated theoretical studies on the triangular-lattice Hubbard model. These materials are quasi-two-dimensional $\kappa$-($\text{ET}$)$_2X$ systems with several possible monovalent anions $X$. ET denotes the bis(ethylenedithio)-tetraphiafulvalene molecule, also often represented as BEDT-TTF, and dimerized pairs of ET molecules constitute a triangular lattice. Each pair provides one conduction electron, and the system is well described by a half-filled Hubbard model on the triangular lattice with nearest-neighbor hopping terms,

$$H = \sum_{\langle i,j \rangle} \sum_{\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} - \mu \sum_{i,\sigma} n_{i\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}. \quad (1.5)$$

Here $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ and, because this model lacks electron-hole symmetry, the chemical potential $\mu$ is introduced to adjust the electron density to half filling, $\sum_\sigma \langle n_{i\sigma} \rangle = 1$. The organic ET compounds have in fact only intermediate correlation strengths: because each site in the model represents a pair of molecules and the corresponding Wannier wave function is extended over the size of this pair, the Coulomb repulsion $U$ is smaller than in the case of typical inorganic compounds, and as a result charge fluctuations have important effects. Because of the non-spherical shape of the molecule pairs, there are two different hopping integrals between nearest-neighbor sites, $t$ and $t'$ [Fig. 1.13(a)]. The ratio $t'/t$ depends on the anion species $X$, and is an important parameter controlling the frustration.

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Extensive investigation of the ET systems [38–40] has demonstrated that their low-energy magnetic properties change dramatically for different anions $X$. Particular highlights in the series include spin-liquid-like behavior in $\kappa$-(ET)$_2\text{Cu}_2\text{CN}_3$ [39] and a reentrant metal-insulator transition with decreasing
temperature at intermediate pressures in \(\kappa-(ET)_2Cu[N(CN)_2]Cl\) [40]. The difference between these systems lies in the different values of the frustration parameter \(t'/t\). Quantum chemistry calculations estimate that \(t'/t = 1.06\) for \(\kappa-(ET)_2Cu_2CN_3\) and \(0.75\) for \(\kappa-(ET)_2Cu[N(CN)_2]Cl\) [38]. From the viewpoint of their electronic structure, the candidate spin-liquid material \(\kappa-(ET)_2Cu_2CN_3\) is very close to being a regular triangular system, in which all the hopping integrals are the same, while the reentrant material \(\kappa-(ET)_2Cu[N(CN)_2]Cl\) corresponds to a triangular geometry perturbed towards an unfrustrated square lattice.

Figure 1.11 shows the temperature-pressure phase diagrams of these two compounds. In these systems, the primary effect of applying pressure is to increase the hopping integrals, and thus the region of higher pressure in experiments corresponds to smaller values of \(U/W\) in the Hubbard model. The boundaries between the metallic and insulating phases in the two materials differ qualitatively in shape: in \(\kappa-(ET)_2Cu_2CN_3\), the insulating phase appears on the high-temperature side of the boundary; in \(\kappa-(ET)_2Cu[N(CN)_2]Cl\), this is only the case above approximately 30 K, while below this the insulating phase extends as the temperature decreases. The former type of behavior is beyond the naive expectation that electron dynamics become more coherent with decreasing temperature, but it is consistent with the conventional Mott transition, as we explain in detail in the next subsection.

![Phase diagram of the Hubbard model determined by dynamical mean-field theory](image)

**Fig. 1.12.** Phase diagram of the Hubbard model determined by dynamical mean-field theory: (a) single-site approximation and (b) results obtained when considering the possibility of AF long-range order, corresponding to an unfrustrated lattice. The dotted line marks the metal-insulator transition when magnetic order is absent. The same acronyms are used for labelling the phases as in Fig. 1.11. (c) Schematic illustration of the electron spectral function at the Mott transition, shown for different values of \(U/W\) (1 \(\rightarrow\) 4).

### 1.4.3 Mott transition in the triangular-lattice Hubbard model

A schematic phase diagram of the “frustrated” Hubbard model is shown in Fig. 1.12(a). There is a first-order phase transition separating metallic and insulating phases. As \(U/W\) is increased, spectral weight is transferred from
the region around $\omega = 0$ to the lower and upper Hubbard bands at $\omega \sim \pm U$ [Fig. 1.12(c)]. The central peak disappears at the transition point and the insulating phase is on the high-temperature side of the boundary. The model is frustrated in the sense that these results are obtained from a single-site, dynamical mean-field theory [41], which assumes the absence of a magnetic instability and thus describes well the case of strong frustration; the phase diagram shows a line of Mott transitions in the original sense of this term, meaning transitions occurring with no simultaneous magnetic order. In the insulating phase, each site has a finite static magnetic moment, which is effectively decoupled from the surrounding moments, and this leads to a large spin entropy of order $\log 2$. This is the reason that the insulating phase is stabilized at higher temperatures. In unfrustrated systems, as shown in Fig. 1.12(b), the metal-insulator transition takes place simultaneously with the emergence of AF long-range order, and the genuine Mott transition does not occur.

![Fig. 1.13.](image)

Fig. 1.13. (a) Generalized triangular lattice with two types of nearest-neighbor hopping integrals, $t$ and $t'$. The case $t = t'$ corresponds to the regular triangular lattice. (b) Phase diagram of the anisotropic, triangular lattice with $t' = 0.8t$.

We now return to the mysterious reentrant metal-insulator transition in $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Cl. This problem was investigated in Ref. [42] by studying the half-filled Hubbard model on an anisotropic, triangular lattice [Fig. 1.13(a)] with $t' = 0.8t$, to determine the $U$-$T$ phase diagram using cellular dynamical mean-field theory (CDMFT). This method is a generalization of the conventional dynamical mean-field theory which uses a cluster of multiple sites (four for this system) [43], and allows one to calculate electronic Green functions and different correlation functions in addition to thermodynamic quantities at finite temperatures. The CDMFT approach has the advantage that both quantum and thermal fluctuations, and thus frustration effects, are taken into account completely inside the cluster.

The phase diagram of the anisotropic, triangular-lattice Hubbard model is shown in Fig. 1.13(b). The band width at $U = 0$, $W = 8.45t$ is taken as the unit of energy. The phase diagram was determined by analyzing the double occupancy $D \equiv \langle n_{i\uparrow}n_{i\downarrow} \rangle$, which is a measure of metallicity. In the metallic
and insulating phases identified in this way, $D(T)$ decreases with decreasing temperature in the paramagnetic insulating (PI) phase, while it increases in the paramagnetic metallic (PM) phase. In the high-temperature regime, these two phases merge smoothly at the crossover line (dotted), which is defined by the condition $dD/dT = 0$. In the low-temperature regime, the two phases are separated by a first-order transition, where the double occupancy shows an abrupt jump.

Fig. 1.14. Wave vector-dependent electron spectral functions of the half-filled Hubbard model on an anisotropic, triangular lattice with $t' = 0.8t$ and $U/W = 0.947$.

It is to be noted that reentrant behavior of the metal-insulator transition/crossover is indeed found in the anisotropic, triangular-lattice Hubbard model for intermediate values, $t'/t \sim 0.8$, of the hopping ratio. The nature of this reentrant behavior is clearly visible in the wave vector-dependent electron spectral function $A_k(\omega)$, shown in Fig. 1.14 for three representative temperatures and at the fixed value $U/W = 0.947$. In the high-temperature PI phase, there is a wide Hubbard gap in the spectrum around $\omega = 0$. In the intermediate PM phase, a heavy-quasiparticle band emerges in the Hubbard gap, a clear sign of metallic behavior and consistent with the conventional Mott transition depicted in Figs. 1.12 (a) and (c). However, the low-temperature, first-order transition line has a different character: the heavy-quasiparticle band does not disappear, splitting instead into two bands separated by a small energy gap, as shown in Fig. 1.14(c). This behavior is similar to the case of a metal-insulator transition driven by magnetic instability.

This type of explanation is confirmed by calculations of the magnetic susceptibility, $\chi_q$ [44], which has a peak at the incommensurate wave vectors $Q \approx \pm (0.7\pi, 0.7\pi)$. This peak grows as the temperature decreases, and diverges at a finite temperature indicated by the crosses in the phase diagram of Fig. 1.13(b). The line of magnetic instability is very close to the first-order metal-insulator-transition line, and it is reasonable to expect that the metal-insulator transition is driven by enhanced magnetic fluctuations. It should also be noted that the two lines are separate and there exists a finite region of a paramagnetic insulating phase between them.

To summarize this section, the anisotropic, triangular-lattice Hubbard model has a phase diagram showing a reentrant metal-insulator transition.
This phenomenon is a direct consequence of the effects of geometrical frustration on magnetic correlations. Taking increasing pressure to reduce the ratio $U/W$, the calculated phase diagram reproduces qualitatively the essential features of the phase diagram of $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Cl. The finite values of the magnetic transition temperature in this 2D model are a consequence of the mean-field-type approximation made for inter-cluster correlations, but may provide an estimate of the true values which would be obtained on including the 3D couplings present in real materials.

1.5 Ordering phenomena at commensurate fermion densities on frustrated geometries

In the preceding sections we have discussed only correlated systems at or near half-filling (i.e. with one electron per lattice site). However, repulsive interactions with longer range than the on-site terms considered above can also give rise to insulating behavior at different commensurate densities. Examples include quarter-filling and even 1/8-filling on the checkerboard lattice, and we illustrate this phenomenon here by discussing two scenarios occurring on frustrated geometries. One is a Bond-Order-Wave (BOW) instability is driven directly by particular nesting properties of the Fermi surface. The other concerns the effects of nearest-neighbor interactions sufficiently strong that they produce a novel type of Mott insulator exhibiting an exotic VBC order. The properties of this Mott insulator may be described by an effective QDM, hence providing a formal connection with Sec. 1.3. This latter insulator can also be doped, a point we mention briefly as a possible route towards quite new and exotic metallic and superconducting behavior.

1.5.1 Bond Order Waves from nesting properties of the Fermi surface

Let us consider the extended Hubbard Hamiltonian, $H = H_0 + H_{\text{int}}$, on the 2D frustrated kagome and checkerboard lattices. We recall here that these lattices are composed of corner-sharing units (respectively triangles and tetrahedra) residing on an underlying bipartite lattice (respectively hexagonal and square), a point which will be important in determining their behavior. The kinetic part of the Hamiltonian is given by

$$H_0 = -t \sum_{\langle ij \rangle} \sum_{\sigma = \uparrow, \downarrow} (c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.}),$$

(1.6)

with positive hopping matrix element $t$, and the sum $\sum_{\langle ij \rangle}$ is over all bonds on the lattice. The interaction part is given by

$$H_{\text{int}} = U \sum_i n_{i\uparrow} n_{i\downarrow} + J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + V \sum_{\langle ij \rangle} n_{i\uparrow} n_{j\downarrow},$$

(1.7)
with on-site repulsion $U$, nearest-neighbor spin exchange $J$, and nearest-neighbor repulsion $V$; in this subsection, we consider the regime of weak and intermediate couplings.

On kagome and checkerboard lattices, the non-interacting Hamiltonian $H_0$ exhibits a dispersionless (flat) band which, for $t > 0$, lies at the top of the spectrum and plays no role. For the kagome lattice, the band structure is remarkable for the presence of “Dirac cones” positioned exactly at the Fermi level of the $1/3$-filled system ($n = 2/3$) and leading to semi-metallic behavior (also relevant on the 3D pyrochlore lattice). Renormalization-group and numerical techniques [45] have been applied to demonstrate that a spontaneous symmetry-breaking occurs for arbitrarily small interactions in this system: the instability corresponds to a BOW in which the kinetic energy is staggered for neighboring triangular units on the underlying hexagonal lattice. We stress that no charge modulation is present (so that all sites remain equivalent), the BOW breaking only the spatial site-inversion $(180^\circ)$ symmetry, such that up- and down-pointing triangles in Fig. 1.2(c) become different, while translational symmetry is preserved [45].

Fig. 1.15. Bond Order Wave on the checkerboard lattice at $n = 1/2$. The four inequivalent bonds are represented by different colors/line types and by different thicknesses (diagonal and $x/y$ bonds are different). All the sites (black dots) carry the same electron occupancy (1/2 electron on average).

On the checkerboard lattice, a BOW instability appears at quarter-filling ($n = 1/2$). As on the kagome lattice, this BOW (Fig. 1.15) is characterized by two types of (tetrahedral) unit with different kinetic (and exchange) energies. However, in the case of the checkerboard lattice, translation symmetry is broken explicitly, although once again there is no charge order. This symmetry-breaking occurs because of perfect nesting of the square Fermi surface [46]. Alternatively, it can be physically understood as special, local (resonant) states formed on the building units (the crossed plaquettes) when the filling is such that these are preferred; the occupation states of the units can also be considered to differ. This local picture is in fact fully equivalent to the nesting instability of the Fermi surface.
1.5.2 Metal-insulator transitions and frustrated charge order

We consider next the strong-coupling limit, where at $U = \infty$ one obtains the Hamiltonian

$$H_{t,J,V} = \mathcal{P}H_0\mathcal{P} + J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + V \sum_{\langle ij \rangle} n_i n_j,$$

(1.8)

$$= H_{t,J} + V' \sum_{\langle ij \rangle} n_i n_j,$$

(1.9)

where $\mathcal{P}$ is the projection operator enforcing the single-occupancy constraint and $V' = V + J/4$. For $V' = 0$, the strong-coupling Hamiltonian reduces to the conventional $t$-$J$ model.

For simplicity, we restrict our considerations to the checkerboard lattice and state only that similar behavior can be found for the kagome lattice. Examining first the limit $V = \infty$ for the special, commensurate filling $n = 1/4$ (1/8-filling), the minimum “classical” energy ($E = 0$) is obtained for all configurations fulfilling the “ice rule” of precisely one particle on every tetrahedron, as in Fig. 1.8(b). The full Hilbert space is then obtained from all possible ways of “decorating” all dimers with a spin index, i.e. the Hilbert space at $n = 1/4$ is exactly that of a two-color QDM [47]. A similar procedure, decorating the simple loop configurations of Fig. 1.8(a), can also be employed to construct the two-color loop configurations which constitute the constrained Hilbert space at quarter-filling ($n = 1/2$) in the large-$V$ limit. It is then clear that, for these special fillings, the GS should be insulating at sufficiently large $V$. The effective dimer (or loop) dynamics can be obtained by perturbation in $t/V$. We note that in the original derivation [33], for spinless fermions, the lowest-order processes were of third order, whereas when spin degrees of freedom are included, terms of dimer-exchange type (below) arise at second order in $t$. Although the constrained quantum dynamics of fermions without [33] and with [48,49] spin differ, the phase diagrams of these models contain a rich variety of crystalline phases, breaking lattice translational and/or rotational symmetry, in both cases. We postpone to Sec. 1.5.3 a discussion of the properties of this type of system away from commensurate filling, and remark only that, among the novel phenomena which can arise, one of the more exotic is the fractionalization under some conditions of a single doped charge $e$ into two $e/2$ components [33,50,51].

Let us now focus in more detail on the insulating phases and consider the effective Hamiltonian acting within the constrained Hilbert space as second-order processes preserving the ice rule. Here $\hat{H} = H_\bullet + H_J$ with

$$H_\bullet = -t_2 \sum_s P_\bullet(s),$$

(1.10)

$$P_\bullet(s) = \left(c_{i_1}^+ c_{j_1}^+ - c_{i_1}^+ c_{j_1}^+ \right) \left(c_{k_1} c_{l_1} - c_{k_1} c_{l_1} \right)$$

$$+ \left(c_{k_1}^+ c_{l_1} - c_{k_1}^+ c_{l_1} \right) \left(c_{i_1} c_{j_1} - c_{i_1} c_{j_1} \right),$$

(1.11)
where \( t_2 = \frac{2t^2}{\epsilon} \) and the index \( s \) labels the empty plaquettes of the checkerboard lattice (Fig. 1.2); the sites of a plaquette \( s \) are ordered as \( i, k, j, l \) in a clockwise (or anti-clockwise) direction. The operator \( P_s(s) \) acts on two electrons forming a singlet bond on one of the two diagonals of \( s \), to rotate this bond by 90 degrees. Candidate GSs breaking the translational symmetry of the lattice are shown in Fig. 1.16. For a quarter-filled band \((n = 1/2)\), the Resonating-Singlet-Pair Crystal (RSPC) of Fig. 1.16(a) was shown to be stable for \( J/t_2 < 1 \) \([48, 49]\). In the twofold-degenerate RSPC, electron pairs resonate on every second empty plaquette, breaking translational symmetry. One therefore expects, on increasing \( V/t \) and \( U/t \), a first-order transition from the BOW state discussed above \([46]\) to the RSPC. The same analysis also provides evidence \([49]\) that the system exhibits plaquette order of the RSPC type also at \( n = 1/4 \) or \( n = 3/4 \), albeit with a quadrupling of the lattice unit cell (as opposed to the doubling found for \( n = 1/2 \)) and a fourfold-degenerate GS. Qualitative differences between these models and their bosonic analogs, which are known for example to exhibit columnar order at \( n = 1/4 \) \([25]\), emphasize the important role of the spin degrees of freedom, not least in stabilizing plaquette phases over phases breaking the rotational symmetry. However, the possibil-

Fig. 1.16. Schematic representation of candidate plaquette (a) and columnar (b) phases of the checkerboard lattice for electron densities \( n = 1/4, 1/2, \) and \( 3/4 \), as discussed in subsection 1.5.2. Dots, shaded plaquettes, and thick (red) lines correspond respectively to electrons, singlet pairs resonating on a plaquette, and resonating four-electron plaquette singlets. From Ref. [49].
ity is being investigated [52] that mixed columnar-plaquette phases, similar to one discovered recently in the square-lattice QDM [23] and which break both \(\pi/2\) rotational symmetry (as does the columnar phase) and translational symmetry in two perpendicular directions (as does the plaquette phase), could be stable in some simple and natural extensions of the Hamiltonian (1.10).

1.5.3 Away from commensurability: doping the Resonating-Singlet-Pair Crystal

Whether plaquette ordering of the RSPC type can survive at sufficiently low but finite hole (electron) dopant concentrations \(x = 1/2 - n\) \(x = n - 1/2\) remains unsettled. It has been shown [48] that phase separation, a generic feature of correlated systems in the vicinity of a Mott phase, is restricted to low hole kinetic energies, meaning to small \(t/J\) and \(t/t_2\) ratios, which leaves an extended regime over which unconventional superconducting pairing may occur. However, the phase diagram of these model for arbitrary electron densities remains largely unexplored, and can be expected to harbor further surprises.

1.6 Summary

The richness and diversity of systems of doped, frustrated magnets and of itinerant correlated electrons on frustrated lattices have been illustrated on selected didactic examples. The dynamics of a small number of doped holes has been investigated in the 2D kagome and checkerboard Heisenberg quantum antiferromagnets revealing striking differences attributed to the different nature of their non-magnetic GS. Two classes of (weakly) doped QDMs have also been discussed, which differ in the assumed statistics, bosonic or fermionic, of the bare holes. We have proceeded further by considering strongly correlated electrons on frustrated triangular lattices, and discuss the physics of an unconventional, reentrant metal-insulator transition. Assuming that increasing pressure reduces on-site correlations, the calculated phase diagram reproduces qualitatively the essential features of the phase diagram of the molecular solid \(\kappa-(ET)_2\text{Cu[N(CN)]}_2\text{Cl}\). As our final example, we consider correlated fermions moving on frustrated lattices at special, commensurate densities for which exotic insulating ground states (GSs) are stabilized. Interesting similarities with frustrated Heisenberg magnets showing an extensive degeneracy of the classical GS manifold are outlined and are shown to be at the heart of their fascinating properties.

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