Doped Singlet-Pair Crystal in the Hubbard model on the checkerboard lattice

Didier Poilblanc,1 Karlo Penc,2 and Nic Shannon3

1Laboratoire de Physique Théorique, C.N.R.S. & Université de Toulouse, F-31062 Toulouse, France
2Research Institute for Solid State Physics and Optics, H-1525 Budapest, P.O.B. 49, Hungary
3H.H. Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol BS8 1TL, UK

(Dated: March 23, 2022)

In the limit of large nearest-neighbor and on-site Coulomb repulsions, the Hubbard model on the planar pyrochlore lattice maps, near quarter-filling, onto a doped quantum fully packed loop model. The phase diagram exhibits at quarter filling a novel quantum state of matter, the Resonating Singlet-Pair Crystal, an insulating phase breaking lattice symmetry. Properties of a few doped holes or electrons are investigated. In contrast to the doped quantum antiferromagnet, phase separation is restricted to very small hopping leaving an extended “window” for superconducting pairing. However the later is more fragile for large hopping than in the case of the antiferromagnet.

PACS numbers: 75.10.-b, 75.10.Jm, 75.40.Mg, 74.20.Mn, 71.10.Fd

Correlated fermions and bosons on frustrated lattices exhibit fascinating properties. For example, on the triangular lattice, hard-core bosons with nearest neighbor (NN) repulsion can realize a supersolid phase exhibiting both charge order and superfluidity [1]. In the light of recent STM experiments suggesting the coexistence of superconductivity and charge order [2], it is interesting to ask whether a fermionic analog of a supersolid could be realized by doping a Mott insulator whose ground state spontaneously breaks lattice symmetries?

In this Letter we study correlated fermions on the checkerboard lattice, a two-dimensional array of corner-sharing tetrahedra. This is a two-dimensional analog of the pyrochlore lattice found in numerous spinel and pyrochlore materials [3]. For a filling of exactly half an electron per site, extremely strong on-site and NN repulsion select a macroscopically degenerate manifold of low-energy configurations fulfilling the so-called “ice rules” constraint, i.e. having exactly two particles per tetrahedron. These can be mapped onto the configuration space of a 6-vertex model [4]. By associating each particle with a dimer joining the centers of the two corner-sharing tetrahedra, an alternative fully packed loop representation can be obtained. Although the constrained quantum dynamics of bosons [3] and spinless fermions [6] differ, the phase diagrams of these models contain a rich variety of crystalline phases breaking lattice translations and/or rotations. In addition, fractionalization of a doped charge e into two e/2 components can also appear under some conditions [6, 7, 8].

Here, we consider the realistic case of spinful fermions (electrons) whose spin degrees of freedom play a central role. We show that the quarter–filled ground–state (GS) is a new insulating crystalline quantum state involving resonant spin singlet electron pairs. We then argue, on the basis of numerical calculations, that pairing can emerge under light doping. Such a superconductor is expected to break lattice symmetries in a similar way to the supersolid of Ref. [1] (although with no charge ordering).

One of the most exciting applications of these ideas is to assemblies of cold atoms in optical lattices, where models of the form we propose can in principle be realized by the appropriate tuning of atomic interactions [8].

The model: Our starting point is a (fermionic) Hubbard model extended with nearest neighbor (⟨ij⟩ in the sum) repulsion V on the checkerboard lattice:

$$H = -t \sum_{\langle ij \rangle} (f_{i\sigma}^\dagger f_{j\sigma} + h.c.) + V \sum_{\langle ij \rangle} n_i n_j + U \sum_i n_i^\dagger n_i \tag{1}$$

using standard notation. We are interested in the limit where the on–site repulsion U is very large, forbidding double occupancy. Furthermore, we consider the limit $V \gg t$, in which case, for quarter filling ($\langle n_i \rangle = \frac{1}{2}$), the ground state is an insulator. Expanding about this state we obtain, to leading order, the effective Hamiltonian:

$$H_\Box = -t_2 \sum_{\square} (f_{i\uparrow}^\dagger f_{j\uparrow} - f_{i\downarrow}^\dagger f_{j\uparrow})(f_{k\downarrow} f_{l\uparrow} - f_{k\uparrow} f_{l\downarrow}) + h.c. \tag{2}$$

where $t_2 = \frac{V^2}{U}$ and the summation runs over the “empty squares” of the checkerboard lattice, with the sites of a given square counted $ijkl$. $H_\Box$ acts on two electrons forming a singlet on the diagonal kl, transferring that singlet to the (empty) perpendicular diagonal ij.

We also introduce a diagonal term which counts the squares where $H_\Box$ can act

$$H_W = W \sum_{\square} \left(\frac{1}{2} - 2S_i \cdot S_j\right) n_i n_j (1 - n_i)(1 - n_k) \tag{3}$$

For $W \equiv t_2$ the Hamiltonian $H_\Box + H_W$ becomes a sum of projectors, and we recover the physics of the Quantum Dimer Model [10] at the Rokhsar–Kivelson point: the GS can be written exactly as an equal-weight superposition of all zero-energy configurations [10]. This nontrivial property also holds in the presence of static holes ($t = 0$).
Finally, we also include the two-site spin exchange

\[ H_J = -J \sum_{\langle ij \rangle} \left( \frac{1}{4} - S_i \cdot S_j \right) n_i n_j , \tag{4} \]

where, for \( U \gg V \gg t \), the exchange constant is given by \( J = \frac{4t^2}{U} + \frac{8t^3}{V} + \frac{16t^3}{U(V-W)} + \frac{16t^3}{U(V-W)} \). For the undoped system \( J \) and \( t_2 \) are therefore independent parameters. Here we consider only antiferromagnetic \( (AF) \) coupling, \( J > 0 \), although a ferromagnetic coupling could also be realized for \( t < 0 \).

Away from quarter filling, when \( N_h \) holes (or electrons) are introduced, simple counting shows that exactly \( 2N_h \) tetrahedra (named hereafter “half-hole tetrahedra”) should contain a single, shared electron (or 3 shared electrons for electron doping). All other tetrahedra contain exactly two shared electrons, as before; violation of this constraint would lead to an energy increase of order \( V \).

The single hopping term \( t \) becomes then effective by moving around the locations of these “fractional charges” \( \frac{e}{2} \). We have performed extensive Lanczos Exact Diagonalization (ED) on a periodic \((45^\circ\) tilted) square cluster of \( N = 32 \) sites in the insulator and for a small number of doped holes or electrons \( [11] \).

Phase diagram at quarter-filling: For \( J = 0 \), by analogy with Ref. \( [3] \) and from simple analytic arguments, we expect to have as a function of \( W \): (i) for large negative \( W \), parallel spin chains; (ii) for intermediate \( |W| \) values a Resonating Singlet-Pair Crystal (RSPC); (iii) for \( W = t_2 \), a liquid-like “RK” point; (iv) for \( W > t_2 \) a manifold of isolated states (which include all ferromagnetic states), all having \( E = 0 \). This picture is indeed supported by our ED calculations (see Fig. (a)) where the characterization of the various phases (see Fig. (b)) can be obtained from the analysis of the low-energy spectrum. In the two fold degenerate RSPC, electron pairs resonate in every second void plaquette, breaking translational symmetry. This is seen numerically in the collapse of a \( k = (\pi, \pi) \) symmetry \( [12] \) singlet excited state onto the GS [Fig. (a)]. For sufficiently negative \( W \) the electrons order along diagonal chains to optimize the Heisenberg exchange along the diagonals of the empty squares, hence breaking rotation symmetry. This is seen in the ED spectrum as the quasi-degeneracy of the \( (k = 0, \pi) \) singlet and the GS. For \( W > 1 \), isolated states where the plaquette resonance is ineffective (including the horizontal chains shown), are favored.

Note that a perturbative expansion about isolated void plaquettes gives a very accurate estimate of the RSPC GS energy (for \( J = 0 \)). Already in first order \( E^{(1)} = N \left( \frac{V}{2} Wa - \frac{t_2}{2} \right) \) as shown in Fig. (a) — the agreement is excellent. A comparison to a similar expansion from disconnected diagonal chains gives an estimate of the boundary between the two phases in full agreement with the numerics, although it suggests that the actual critical value shifts to more negative \( W \) in the thermodynamic limit. One should point out that (i) in contrast to the bosonic case \( [2] \), the range of stability of the plaquette phase is much broader on the \( W \) axis and (ii) as shown in Fig. (a) and Fig. (b) the RSPC is also very stable when the exchange \( J \) is included. Only above \( J/t_2 \sim 1.5 \), a new phase of 4-electron plaquettes oriented along the \((1, \pm 1)\) directions is stabilized by a gain of exchange energy (small Heisenberg loops have higher quantum fluctuations) \( [13] \). Lastly, we stress that our RSPC at \( J/t_2 < 1.5 \) differs from the Bond Order Wave realized for \( t > 0 \) at intermediate \( V/t \) ratio (for which charge fluctuations still occur) \( [14] \) which shows two kinds of “criss-crossed” plaquettes. Indeed, although both phases exhibit a doubling of the unit cell (neither with charge ordering), they differ in the type of resonating plaquettes.

Finite doping: In investigating the effect of doping, we restrict to \( W = 0 \) and consider only values of \( J \) for which the RSPC is realized at zero doping.

A single doped hole (or electron), as discussed above, is in fact split into two mobile “half-hole tetrahedra” carrying one electron only (and effective charge \( e/2 \)) \( [6, 7] \). For reasons similar to those given in Ref. \( [6] \), one expects a confining potential so that a small quasiparticle weight survives (in fact, in the \( t = 0 \) limit, the two “half-hole tetrahedra” even share a common site). In order to estimate the coherent bandwidth we have computed by ED the GS energy of a single hole for various inequivalent \( k \) momenta \( [15] \) and results are reported in Fig. (a). For large \( t \) (compared to \( t_2 \) or \( J \)) the bandwidth is reduced by, roughly, a factor \( t_2/|t| \) (or \( J/|t| \)) for \( t > 0 \), or even more for \( t < 0 \), and the hole becomes quite massive. This is very reminiscent of the behavior of a single hole doped in...
a quantum antiferromagnet (shown also in Fig. 3 for comparison) where the hole leaves behind a path of flipped spins which can be healed over a short time scale \( \propto 1/J \) by spin fluctuations \( [16] \). We believe a similar path along which the plaquette order is perturbed exists also here behind the moving hole.

As the effective Hamiltonian lowers the energy of singlet pairs, it is natural to study the emergence of pairing interaction. For \( t = 0 \) we found that two holes sit next to each other or, more precisely, that all four “half-hole tetrahedra” are fully packed around a single void plaquette in order to minimize plaquette resonance and bond exchange energies. The two-hole GS for \( t = 0 \) can be very well approximated by \( |\Psi_{2h}\rangle \simeq \Delta_l |\Psi_0\rangle \) where \( \Delta_l \) removes two electrons along the diagonals of a void plaquette with \( s \)-wave orbital symmetry. For finite \( t \) it is therefore convenient to define the overlap squared \( Z_{2h} = |\langle \Psi_{2h}|\Psi_{2h}\rangle|^2 \), to get an accurate estimate of the so-called 2-hole Z-factor \( |\langle \Psi_{2h}|\Delta_l^2|\Psi_0\rangle|^2 \). Note that \( |\Psi_{2h}\rangle \) is now defined as a Bloch state with the same momentum as the 2 hole GS \( |\Psi_{2h}\rangle \) (typically \( k = (0,0) \)) i.e. as a linear superposition of all local (degenerate) hole pair states. Increasing \( |t| \), \( Z_{2h} \) should remain finite as long as the holes stay bound. Fig. 4 shows that \( Z_{2h} \) is more rapidly suppressed than for two holes in an AF \( [18] \) (shown also in Fig. 4 for comparison), suggesting that the hole pair is more fragile here for large \( |t|/t_2 \) and \( |t|/J \) values. A very similar behavior is also found for two doped electrons (see Fig. 4) although the related bound state has now \( d \)-wave symmetry \( [17] \).

To get a more direct insight on the tendency of a hole pair to break up at large \( |t|/t_2 \) we have computed the pair binding energy \( \Delta_{2h} = E_{2h} + E_{0h} - 2E_{1h} \) shown in Fig. 5 where \( E_{mh} \) is the GS energy of the system with \( N_h = m \) holes. From its definition, negative \( \Delta_{2h} \) suggests the binding of the holes, as previously seen in the doped Heisenberg AF on the square lattice (in blue) — even for rather large hole kinetic energy. On the contrary, the data for the \( 1/4 \)-filled checkerboard lattice do not provide evidence of binding when \( t/t_2 > 1 \). Note however that \( \Delta_{2h} \) is subject to stronger finite size effects than \( Z_{2h} \), so a weak pairing might still survive for \( |t|/t_2 > 1 \).

Since phase separation might compete with superconductivity, one needs now to consider the four-hole binding energy, \( \Delta_4 = E_{4h} + E_{0h} - 2E_{2h} \), from which an estimate of the compressibility \( \kappa = \frac{\partial^2 E}{\partial n_h^2} \) at small hole density \( n_h = \frac{N_h}{N} \) can be given using \( \kappa^{-1} \simeq N\Delta_4 \). Phase separation signaled by a negative curvature of \( E(n_h) \) implies, in this case, the formation of four-hole droplets (quartets) i.e. \( \Delta_4 < 0 \). Our numerical calculation of \( \Delta_4 \) on 32 sites shows that quartets are stable when \( t \) is small
enough, the plaquette resonance and bond exchange providing an effective “glue” to bind holes together. Such a behavior is similar to the case of the doped quantum AF [19]. However, here a very small hopping $t$ can easily suppress phase separation while affecting pairing only slightly. Indeed, for a typical value of $J/t_2 = 0.5$, $\Delta_4 \simeq -0.0635 t_2$ (meaning $\kappa < 0$) for static holes ($t = 0$) while $\Delta_4 \simeq -0.0042 t_2$ ($\Delta_4 \simeq -0.00111 t_2$) for $t = 0.1 t_2$ ($t = -0.1 t_2$) and $\Delta_4 \simeq 2.471 t_2$ ($\Delta_4 \simeq 2.044 t_2$) for $t = 0.5 t_2$ ($t = -0.5 t_2$). We therefore expect a paired state when $t/t_2 > 0.1$.

Lastly, we would like to draw possible scenarios at finite doping: (i) If plaquette ordering survives at sufficiently small doping (plausible since the RSPC is “protected” by a finite gap), one expects a superconducting phase which inherits broken translational symmetry from its parent Mott insulator (the RSPC). This can be viewed as a new type of supersolid with no charge modulation [20]; (ii) Alternatively, hole pairs could arrange along domain walls between out-of-phase RSPC regions. This scenario is however unlikely, providing no obvious gain of kinetic energy w.r.t the supersolid.

To summarize, on the checkerboard lattice, at quarter-filling (one electron per two sites) and in the limit of large NN repulsion (enforcing the “ice rule” constraint on every tetrahedron), the Hubbard model exhibits a robust insulating Resonating Singlet-Pair Crystal with a uniform average charge. We provide arguments for Cooper pair formation and argue in favor of a supersolid phase in an extended region of parameters, phase separation being confined only at very small hole kinetic energy. However, hole-Cooper-pairs seem to be more fragile for large hopping than in the doped antiferromagnet on the square lattice.

We thank P. Fulde and F. Pollmann for numerous discussions. D.P. thanks the Agence Nationale de la Recherche (ANR) and IDRIS (Orsay, France) for support. K.P. is grateful for the support of the Hungarian OTKA Grants No. T049607 and No. K622800. N.S. acknowledges support under EPSRC Grant No. EP/C539974/1.

[1] S. Wessel and M. Troyer, Phys. Rev. Lett. 95, 127205 (2005); D. Heidarian and K. Damle, Phys. Rev. Lett. 95, 127206 (2005); R.G. Melko, A. Paramekanti, A.A. Burkov, A. Vishwanath, D.N. Sheng, and L. Balents Phys. Rev. Lett. 95, 127207 (2005).
[2] T. Hanaguri et al., Nature 430, 1001 (2004).
[3] “Frustrated Spin System”, Ed. H.T. Diep, World Scientific (2004).
[4] E.H. Lieb, Phys. Rev. Lett. 18, 1046 (1967). For recent attempts to quantize vertex models, see E. Ardonne, F. Fendley, and E. Fradkin, Ann. Phys. 310, 493 (2004); O. F. Syljuasen, and S. Chakraarvty, Phys. Rev. Lett. 96, 147004 (2006).
[5] N. Shannon, G. Misguich, K. Penc, Phys. Rev. B 69, 220403(R) (2004); see also J.-B. Fouet, M. Mambro, P. Sindzingre, and C. Lhuillier, Phys. Rev. B 67, 054411 (2003) for the Heisenberg model.
[6] F. Pollmann, J.J. Betouras, K. Shtrengel, and P. Fulde, Phys. Rev. Lett. 97, 170407 (2006); F. Pollmann, P. Fulde, and E. Runge, Phys. Rev. B 73, 125121 (2006).
[7] P. Fulde, K. Penc and N. Shannon, Annales der Physik, 11, 892 (2002).
[8] In general, the two components are bound by a shallow “string” potential.
[9] J.H. Büchler et al., Phys. Rev. Lett. 95, 040402 (2005).
[10] D.S. Rokhsar and S.A. Kivelson, Phys. Rev. Lett. 61, 2376 (1988).
[11] For two holes (two electrons) doped, the GS $k = 0$, $A_1$ ($B_1$) and Time-Reversal $TR = -1$ irreducible representation contains 9 244 689 (130 840 618) states.
[12] A reduced Brillouin zone (BZ) scheme is used (two-site unit cell). The states are classified according to the $A_1$, $A_2$, $B_1$, $B_2$, and $E$ irreducible representation of the point group $D_4$ (origin at the center of an empty square).
[13] For $W > 0$ ($W < 0$) an out-of-phase (in-phase) arrangements of these diagonal lines of 4-electron plaquettes is realized to maximize (suppress) $t_2$ resonances.
[14] M. Indergand, C. Honerkamp, A. Läucli, D. Poilblanc and M. Sigrist, Phys. Rev. B 75, 045105 (2007).
[15] All momenta along the line $k_x = 0$ as well as $k = (\pi, \pi)$ have been considered (reduced BZ).
[16] D. Poilblanc, T. Ziman, H.J. Schulz, and E. Dagotto, Phys. Rev. B 47, 14267 (1993) and references therein.
[17] For e.g. $|t| = t_2$ and $J/t_2 = 0.5$, the 2-doped electron (2-hole) GS energies are $-27.428$ $(-25.935)$ and $-24.704$ $(-23.220)$, in units of $t_2$, for $t \geq 0$ and $t < 0$ respectively.
[18] D. Poilblanc, Phys. Rev. B 48, 3368 (1993); A.L. Chernyshev, P.W. Leung, and R.J. Gooding, Phys. Rev. B 58, 13594 (1998).
[19] D. Poilblanc, Phys. Rev. B 52, 9201 (1995).
[20] A supersolid might also be realized near half-filling; See D. Poilblanc, Phys. Rev. Lett. 93, 197204 (2004).