Gapless spin liquids: Stability and possible experimental relevance

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Gapless Spin Liquids: Stability and Possible Experimental Relevance

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For certain crystalline systems, most notably the organic compound $\mathrm{EtMe_3Sb[Pd(dmit)_2]_2}$, experimental evidence has accumulated of an insulating state with a high density of gapless neutral excitations that produce Fermi-liquid-like power laws in thermodynamic quantities and thermal transport. This has been taken as evidence of a fractionalized spin liquid state. In this paper, we argue that *if the experiments are taken at face value*, the most promising spin liquid candidates are a Z_4 spin liquid with a spinon-Fermi surface and no broken symmetries, or a Z_2 spin-liquid with a spinon-Fermi surface and at least one of the following spontaneously broken: (a) time-reversal and inversion, (b) translation, or (c) certain point-group symmetries. We present a solvable model on the triangular lattice with an (a) type Z_2 spin liquid groundstate.

The notion of a "spin-liquid phase" – a quantum disordered insulating phase which is not adiabatically connected to a band insulator – has captured the imagination of theorists for decades [1, 2]. In recent years, several developments have increased interest in this subject [4], including a number of exact results for solvable models which prove the existence of spin liquids as theoretically stable quantum states of matter [5–9], numerical studies [10–12] of more physically realistic models, increasingly sophisticated field theoretic analyses [3, 13], and, importantly, recent experimental results. Specifically, a number of quasi-two-dimensional (2D) insulating materials have been found to exhibit highly unusual low temperature thermodynamic and transport properties which are unlike those expected of conventional phases [4, 14–16].

One of the most notable examples is the organic material $\mathrm{EtMe_3Sb[Pd(dmit)_2]_2}$ (referred to as "dmit"), which provides a physical realization of a frustrated spin-1/2 system on a (anisotropic) triangular lattice. Although dmit has an odd number of electrons per unit cell, the charge response (conductivity) is insulating and NMR studies indicate that no magnetic ordering occurs down to the lowest achievable temperatures (~ 20mK), which are much smaller than the scale of the characteristic exchange coupling, $J \approx 250K$ [18]. The specific heat, C, uniform susceptibility, χ, and thermal conductivity, κ_{xx}, exhibit T dependencies consistent with Fermi-liquid-like power laws, $C \sim k_B^2 \rho T$, $\chi \sim \rho \mu_B^2$, and $\kappa_{xx} \sim C$, with an apparent density of states, $\rho \sim 0.1 J^{-1}$ per unit cell [16, 19]. This is suggestive of the existence of a spin-liquid with a charge gap, and neutral, fermionic spinon excitations with a nonzero density of states at zero energy and an estimated mean free path $\sim 0.5\ \mu m$ [16]. Similar observations in a closely related compound, κ-$\mathrm{(ET)_2Cu_2(CN)_3}$ (referred to as κ-ET)[17], led to the proposal of a candidate spin liquid, with a "pseudo Fermi surface" of spinons and an emergent $U(1)$ gauge field[20, 21]. Eventually it was found that κ-ET has a gap to mobile excitations,[22] unlike the case in dmit.

In this paper, we *assume* that dmit realizes a fractionalized spin liquid state, and we address the problem of identifying the most promising candidates to explain the experiments. In particular, we assume that the experimental claims summarized above can be taken at face value, and we also consider the possibility that quenched disorder plays no fundamental role. In agreement with previous discussions [20], we argue that under these assumptions the $U(1)$ spin liquid is not a viable candidate.

We introduce a Z_2 spin liquid phase with a *stable* spinon-Fermi surface *and* spontaneously broken time-reversal and inversion symmetry as a promising candidate state for dmit. Other attractive candidates with spinon-Fermi surfaces which are only marginally unstable are a Z_4 spin liquid with no necessary broken symmetries, or a Z_2 spin liquid which spontaneously breaks translation symmetry [21], or certain point-group symmetries [45]. For the Z_4 spin liquid, we provide a physical mechanism by which it can arise, through quantum melting a spinon pair-density wave state. Interestingly, we find that a marginally unstable featureless Z_2 spin liquid with a spinon-Fermi surface can exist on the square lattice, but apparently not on the triangular lattice.

We have studied the magnetic field response of these spin liquids, and conclude that these states are compatible with existing magneto-thermal transport measurements. As we will discuss, an attractive feature of our proposals, as compared with some existing ones, is that they do not require an extremely small scale for spinon pairing. The broken symmetries of the candidate Z_2 spin liquids with spinon-Fermi surfaces imply the existence of at least one thermal phase transition.

All known spin liquids are fractionalized phases described, at low energies, by an effective theory consisting of matter ("spinons") coupled to an emergent gauge field. An interesting subset of these states can be thought of as derived from an underlying theory of a fermionic spinon metal interacting with an emergent $U(1)$ gauge field. In analogy with superconductors, the spinons can condense in pairs or clusters, breaking the $U(1)$ gauge symmetry to a discrete subgroup and gapping the gauge-field fluctuations via the Anderson-Higgs mechanism. Recently, it has been recognized [29, 32] that exotic superconducting states can exist with appropriate broken symmetries which support a Fermi surface of Bogoliubov quasi-particles, while still exhibiting the standard Meissner effect; the spin-liquid analogue of these states have a gap to all gauge-field fluctuations in the presence of a robust spinon Fermi surface, and are therefore promising candidates to explain the dmit phenomenology.

"Parent" $U(1)$ spin liquid – The $U(1)$ spin liquid can be understood [3] by representing the spin operator as $\vec{S} = \Psi^\dagger \vec{\underline{\tau}} \Psi$, where Ψ is a two-component spinor field (corresponding to the two polarizations of a spin-1/2 fermion), and $\underline{\tau}_\alpha$ are the Pauli matrices. This leads to a minimal model of a sea of spinons coupled to an emergent $U(1)$ gauge field, given by the Euclidean Lagrangian:

$$L = \Psi^\dagger \left[\partial_t - i a_0 - \underline{\epsilon}(-i\vec{\nabla} - \vec{a}) \right] \Psi + |f|^2/(2g) + \ldots, \quad (1)$$

$\underline{\epsilon} = \epsilon_0 \underline{1} + \sum_\alpha J_\alpha \underline{\tau}_\alpha$ is a 2×2 matrix; time reversal symmetry implies that $\epsilon_0(\vec{k}) = \epsilon_0(-\vec{k})$ and $J_\alpha(\vec{k}) = -J_\alpha(-\vec{k})$, and, in the absence of spin-orbit coupling, spin-rotational symmetry implies $J_\alpha = 0$. Here, $\vec{k}$ signifies the Bloch wave-vector and, where there are multiple spinon bands, it is implicitly assumed to include a band index. $f_{\mu\nu}$ is the field strength corresponding to the emergent gauge field, a_μ, and $\ldots$ represents gauge-invariant four-fermion and higher order interaction terms. L is proposed to describe physics at energies small compared to the charge-gap, so the remaining degrees of freedom carry zero electro-magnetic charge.

The weak-coupling ($g \to 0$) fixed point is unstable. One possible result is a strong-coupling non-Fermi-liquid phase which does not break any symmetries, does not have any well defined quasiparticles, but which preserves the Fermi surface [25]. However, even assuming this to be a stable phase, it cannot be responsible for the experimental claims summarized above. It would exhibit power-laws (for example, $C \sim T^{2/3}$) that are substantially different from those of a Fermi-liquid, unless a broad intermediate finite-temperature regime is assumed that is governed by the unstable ($g \to 0$) fixed point. Given that the gauge coupling is strongly relevant and there are no naturally small parameters in the problem, we consider such a broad intermediate regime unlikely. Additional issues that cast doubt on the $U(1)$ spin liquid below 1 K on the basis of the thermal Hall data were presented in Refs. [16, 26].

Breaking $U(1)$ to Z_2 – The gauge symmetry in Eq. (1) can be spontaneously broken due to pairing of spinons, gapping the gauge fluctuations and leaving a residual Z_2 gauge symmetry. Because a Z_2 gauge theory has no finite temperature transition in two dimensions [27], spinon pairing defines a crossover rather than a phase transition. The discreteness of the residual gauge symmetry implies there are gapped, vortex-like excitations ("visons"[28]) which carry half a quantum of gauge flux.

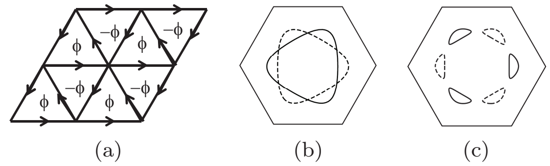

FIG. 1. (a) Loop order on the triangular lattice where ϕ represents the phase accumulated in circling the plaquette. This leads to the dispersion relation $\tilde{\epsilon}(\vec{k}) = -t[\cos(k_x) + \cos(k_+) + \cos(k_-)] - \delta[\sin(k_x) + \sin(k_+) + \sin(k_-)]$ where $k_\pm = -k_x/2 \pm \sqrt{3}k_y/2$ and $\delta/t = \tan(\phi/3)$. (b) The Fermi surface with $\phi = \pi/2$ (solid line) and the same curve with $\vec{k} \to -\vec{k}$ (dashed line). (c) The residual Fermi surface in the presence of a small pairing term.

At energies low compared to the vison creation energy the resulting effective field theory is of the form:

$$\begin{aligned} L = {} & \Psi^\dagger \left[\partial_t + \underline{\epsilon}(-i\vec{\nabla}) + \underline{\delta}(-i\vec{\nabla}) \right] \Psi \\ & + \Psi^\dagger \underline{\Delta}(-i\vec{\nabla}) \Psi^\dagger + \Psi \underline{\Delta}^\dagger(-i\vec{\nabla}) \Psi + \cdots . \end{aligned} \quad (2)$$

The induced changes in the dispersion, $\underline{\delta}(\vec{k})$, can be absorbed into a redefined spinon "band structure," $\underline{\tilde{\epsilon}}(\vec{k}) = \underline{\epsilon}(\vec{k}) + \underline{\delta}(\vec{k})$.

Z_2 spin-liquids with a spinon-Fermi surface – In the $U(1)$ spinon Fermi surface state, arbitrary pairing terms cannot appear in the theory as they are forbidden by the $U(1)$ gauge symmetry. However once the $U(1)$ is broken to Z_2, the spinon number is only conserved mod 2, so any pairing terms can generically appear. Addition of a small s-wave pairing term would open a gap everywhere, while p-wave or d-wave pairing terms will gap the Fermi surface everywhere but at isolated nodal points.

The instability inherent in a state with a spinon Fermi surface with a Z_2 gauge field is removed if the system in question breaks both time-reversal and inversion symmetry. In this case, the degeneracy of states at $\vec{k}$ and $-\vec{k}$ is lifted. (See Fig. 1.) If we draw the Fermi surface corresponding to $\tilde{\epsilon}(-\vec{k})$, (dashed line in Fig. 1b) a small pairing term will open gaps only in the neighborhood of the points at which the two copies of the Fermi surface happen to cross. The rest of the Fermi surface is perturbatively stable!

Therefore, if both time-reversal and inversion symmetry are broken, it is possible to stabilize a Z_2 spin liquid with a spinon-Fermi surface. Such a state was found [9] in a Γ-matrix model on the Kagome lattice with explicit time-reversal and inversion symmetry breaking. A related result was obtained in another context: in [29] it was shown that a Fermi surface occurs in a state with coexisting d-wave superconducting and orbital loop order.[30] An example of such a state on a triangular lattice is shown in Fig. 1. A subtle point is the possibility that the spinon band structure violates time-reversal and inversion, while physical (gauge invariant) quantities preserve these symmetries [31]. However such states are believed to be marginally unstable to translation symmetry breaking that gaps the Fermi surface.

To affirm the possibility of a stable Z_2 state with a spinon-Fermi surface, we introduce (in the Supplemental Material) an exactly solvable version of the Γ-matrix model on the triangular lattice. The model itself is not inversion symmetric. We find that for a range of parameters, the ground-state spontaneously breaks time-reversal symmetry and supports a stable spinon Fermi surface coupled to a Z_2 gauge field. This proves the existence of this state as a quantum phase of matter.

Another possibility is a pair density wave (PDW) state for the spinons. A specific version of this was proposed in [21]. The essential feature [32] of this state which prevents it from fully gapping the spinon-Fermi surface is that the gap parameter changes sign, $\underline{\Delta}(\mathbf{r}) = -\underline{\Delta}(\mathbf{r} + \hat{\mathbf{e}}\lambda/2)$ under translation by 1/2 the PDW period, λ, so that the spatial averaged gap vanishes. ($\mathbf{Q} = (2\pi/\lambda)\hat{\mathbf{e}}$ is the PDW ordering vector.) In the special case where the period is two lattice spacings, the PDW does not actually break translation symmetry, since translation by one lattice constant is equivalent to a uniform gauge transformation. On the triangular lattice such a state does, however, break the C_6 rotational symmetry. However, on a square lattice, a PDW state of $\mathbf{Q} = (\pi, \pi)$ does not break any symmetries. In these cases, the spinon-Fermi surface has a marginally relevant Cooper instability.

Z_4 spin liquids – We can also imagine cases where spinon quartets condense but not pairs, leaving a Z_4 gauge theory with a richer collection of vortex-like modes. While quartet condensation is relatively unnatural for electrons, with their strong repulsive interactions, there is no particular reason to rule out condensation of higher multiplets in the case of spinons. It is, for example, possible to construct model electron problems with strong, spin-dependent attractions which exhibit a charge 4e superconducting phase [33].

If the quartet binding energy Δ_{4e} is large enough, the fermion spectrum will be fully gapped. However if Δ_{4e} is much less than the Fermi energy, it is possible instead for a stable Fermi surface to coexist with the quartet condensate. Treated perturbatively about the Fermi liquid fixed point, a quartet condensate simply introduces four-fermion interactions that conserve charge modulo four; these can only introduce a marginal instability, similar to the usual Cooper instability of the Fermi liquid (see Supplemental Materials for additional details).

One physical way for a charge-4e condensate with Fermi surface to occur is the following. Consider starting with a pair-density wave (PDW) or FFLO state, which are known to support stable Fermi surfaces. If such a state is quantum or thermally melted by the proliferation of double dislocations, then the pair order parameter will be disordered, leaving a residual charge-4e superconducting order [34–36]. At the quantum critical point, the melting of the order parameter only affects the Fermi surface at "hot spots" – points on the Fermi surface that can be connected by integer multiples of the PDW wave vector. Therefore it is clear that most of the Fermi surface will survive the melting transition intact and persist in the presence of the quartet condensate. In the spin-liquid context, such a state would be a Z_4 spin-liquid with a spinon Fermi surface.

A Z_4 spin liquid is an attractive candidate for accounting for the experiments - in contrast to the Z_2 spin liquid, it does not necessitate time-reversal and inversion, translational, or certain point group symmetry breaking to ensure the stability of the spinon-Fermi surface over a broad intermediate energy scale.

Response to magnetic field – Experiments on dmit have reported no measurable thermal Hall angle up to an applied field of 12 T [16], and an interesting upturn in $\kappa_{xx}(T = 0)$ starting at an applied field of 2 T.

The orbital coupling to charge fluctuations leads to a linear coupling between the magnetic field and the spin chirality [38]. The spin chirality is proportional to the magnetic flux of the emergent gauge field [3]. However, the visons of Z_2 spin liquids typically are even under time-reversal, due to tunneling between $\phi_0/2$ and $-\phi_0/2$ vortices. For strong enough tunneling, vortices are therefore not induced by a magnetic field, even if it is larger than the vison gap. If the tunneling amplitude is sufficiently small, an applied magnetic field could mix the symmetric and antisymmetric states, stabilizing vortices relative to anti-vortices.

If the effective penetration length of the spinon condensate is small (i.e. if it forms a Type I superconductor), then we expect that a magnetic field will not induce a finite density of vortices, and therefore there is no mechanism for modifying the thermal Hall effect. If the spinon condensate forms a Type II superconductor, then a magnetic-field dependent thermal Hall effect is possible in principle, although at present we do not have a theoretical estimate of its magnitude.

Assuming the Type I scenario, the only response of the spin liquid to the magnetic field is through the Zeeman coupling. For the spinon-Fermi surface states discussed in this paper, this will change the density of states and will not modify the Hall response. However, for the Z_2 spin liquid with a stable spinon-Fermi surface with spontaneously broken time-reversal and inversion symmetry, there will be a zero-field anomalous thermal Hall response.In the clean limit, the only contribution to the thermal Hall conductivity comes from[40] the Berry curvature of the Bloch states of the spinons: $\kappa_{xy} = \frac{\pi^2}{3}\frac{k_B^2 T}{h}\frac{1}{2\pi}\int d^2k f_{xy}(\vec{k})n(\vec{k})$, where $f_{xy}(\vec{k})$ is the Berry curvature, and $n(\vec{k})$ is the occupation number of the partially filled bands. Generically, we expect: $\frac{1}{2\pi}\int d^2k f_{xy}(\vec{k})n(\vec{k}) \sim 1$. Therefore, we estimate: $\kappa_{xy}/T \sim \frac{\pi^2}{3}\frac{k_B^2}{h} \approx 10^{-12}$ W/K^2 .

In order to compare with the experimental results in dmit, consider κ_{xy}/T per layer, where the layer spacing is $d \approx 1.5$ nm: $\kappa_{xy}/Td \approx 6 \cdot 10^{-4}$ W/K^{2}m. Using the zero temperature intercept of the longitudinal thermal conductivity, $\lim_{T\to 0} \kappa_{xx}/Td = 0.2$ W/K^{2}m, we predict a Hall angle $\tan\theta_H = \kappa_{xy}/\kappa_{xx} \approx 0.003$. The experimental error bars on the Hall angle were reported to be ≈ 0.05 at 0.23 K, ≈ 0.02 at 0.70 K, and 0.003 at 1 K and 12 T [16]. Our prediction is over an order of magnitude less than the experimental error bars on the low temperature data and therefore appears to be consistent with experiment.

The other observed feature in the magneto-thermal transport in dmit is a rapid upturn in $\kappa_{xx}(T = 0)$ at 2 T. One possible explanation is that the tunnel splitting in the vison state is small, so that the gap to vortex formation is approximately $1K$, and hence magnetic fields in excess of 1 T lead to vortex proliferation. However, the vortex state would be expected to exhibit a thermal Hall effect due to scattering of the spinons from vortices, and this has not been observed. It is possible that the experimental error bars are still too large to observe this effect. An alternative possibility is that the upturn in κ_{xx}/T is simply due to variations of the density of states in the spinon band structure.

Since the Z_4 spin liquid with a spinon-Fermi surface need not break time-reversal symmetry, its anomalous thermal Hall response can be exactly zero. The structure of its vortices is richer; the Z_4 visons are not all time-reversal symmetric and will therefore couple linearly to a magnetic field, leading to the possibility of a non-zero thermal Hall effect. However, the same quantitive issues discussed in the Z_2 context apply here, as well.

Discussion – All known spin liquids can be classified in terms of a spectrum of gapless spinons (or their absence) and the nature of the emergent gauge fields to which they couple. In addition to a spinon Fermi surface, nodal fermions, with dispersion $\epsilon(k) \sim k^\alpha$ are another possibility. The familiar case of $\alpha = 1$ has a vanishing density of states at zero energy, and so, it is not a candidate to explain the experiments if the effects of disorder are negligible. It has been noted, however, that weak disorder broadens the nodes, resulting in a constant density of states at zero energy proportional to the spinon scattering rate, $1/\tau$. Thus, only if the disorder plays a significant role in the thermodynamics can a nodal spin-liquid be a candidate to explain the experiments. Moreover, in this case the thermal conductivity is theoretically expected to be "universal" in the sense that it does not depend on τ, although it does depend on the anisotropy of the nodal dispersion; as we will show in the Supplemental material, to account for the magnitude of the observed thermal conductivity, one must assume an extremely large anisotropy corresponding to a pairing scale that is roughly 500 times smaller than J. In contrast, none of the proposed states with spinon-Fermi surfaces require the existence of such an unnaturally small pairing scale. The case of $\alpha = 2$ ("quadratic band touching") [41] would produce a finite density of states, although at the expense of rendering the state marginally unstable in the presence of interactions [42]. More importantly in the present context, naive scaling suggests a thermal conductivity $\kappa_{xx} \sim C\ \overline{v}l \sim T^{3/2}$, where the mean-squared velocity, $\overline{v^2} \sim T$. An interesting possibility is given in [24], which consists of both a spinon-Fermi surface of spin-1 fermionic excitations and a node with dispersion $\epsilon(k) \sim k^3$. This state also breaks time-reversal and inversion, and it further requires weak disorder to exhibit power-laws consistent with experiment.

As we have discussed, the spinon-Fermi surface is *stable* in the Z_2 case only if time-reversal and inversion are broken, and marginally unstable if only translation and/or rotational symmetry is broken.[46] Otherwise, even achieving marginal stability requires a larger gauge group, such as Z_4 [47].

One experimental signature of a chiral Z_2 state is that the breaking of time-reversal symmetry must occur at a finite T transition (presumably in the Ising universality class) [43], and such a transition should be observable in any thermodynamic quantity. Existing specific heat data show no sharp anomaly, but other thermodynamic quantities, such as elastic moduli, might be more sensitive. In the time-reversal breaking phase, various anomalous response functions should be non-zero. For example, as we have discussed, one expects a non-zero anomalous thermal Hall effect, although it may be small. One also expects a spontaneous Kerr effect, which can be measured with exquisite sensitivity [44]. When spin-orbit coupling is taken into account, there should be small magnetic fields which might be detectable in NMR or μSR, although since these fields are proportional both to the magnitude of the time-reversal symmetry breaking order parameter and to the spin-orbit coupling, they may be quite small.

The marginal instability of the time-reversal invariant spin liquids with spinon-Fermi surfaces has possibly interesting experimental implications; it could account for the existence of small energy scales in the problem which could depend sensitively on minor differences between materials. For instance, it might account for the low temperature gap inferred from thermal transport in κ-ET [22] but not in dmit.

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[45] If weak quenched disorder does play a fundamental role, then other spin liquid states may also be viable candidates [23, 24].

[46] The C_6 rotational symmetry of an ideal triangular lattice is weakly broken by the crystal structure of dmit; consequently, what would have been a sharp symmetry

breaking transition would be somewhat rounded.
[47] A Z_3 gauge field is another simple possibility [6, 7], but it does not have a simple slave-particle construction.