How to Measure a Spinon Fermi Surface

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We propose an experiment to identify the potential existence of a spinon Fermi surface by looking for oscillatory coupling between two ferromagnets via a spin liquid spacer. Three candidate spin liquids are investigated, and it is found out that in all cases long period oscillations should be present, the period of which would identify the Fermi wave vector of the spinon surface.

In 1973, Anderson [1] proposed the possibility of a spin liquid, a state where long range magnetic order is suppressed by frustration, low dimensionality and/or quantum fluctuations. In 1987, he resurrected this idea in the context of high temperature cuprate superconductors [2]. The proposed ground state was a so-called uniform resonating valence bond (RVB) state which possesses a Fermi surface for spin excitations. Although it turns out that the undoped cuprates are antiferromagnets, the spin liquid concept in the context of doped cuprates is a very active field of study [3].

Frustration plays a major role in suppressing magnetic order, and the original RVB idea was developed for a triangular lattice. Since then, other frustrated lattices have been identified, including pyrochlore, kagome, and hyperkagome [4]. In the past few years, three candidate $S = 1/2$ spin liquids have been identified [5]: $\kappa - (BEDT - TTF)_2Cu_2(CN)_3$ (distorted triangular lattice) [6], $ZnCu_3(OH)_6Cl_2$ (kagome) [7], and $Na_4Ir_3O_8$ (hyperkagome) [8]. All of these materials are insulating, have a large Curie-Weiss temperature, and yet show no ordering down to the lowest temperatures measured. In each case [9–13], a uniform RVB state has been proposed as the ground state with a resulting spinon Fermi surface. There is indirect evidence for such a surface. The BEDT salt exhibits a linear T specific heat [14] characteristic of a Fermi surface. The herbertsmithite $ZnCu_3(OH)_6Cl_2$ exhibits a sublinear T dependence of the specific heat [15], but this can be understood from self-energy corrections [9]. In addition, a Pauli-like susceptibility is inferred at low temperatures once the effect of impurity spins has been factored out.

On the other hand, this evidence for a spinon surface is indirect. Spin glass behavior, or a magnon dispersion, ω^z, with a power z equal to the dimensionality, can also lead to a linear T specific heat [4]. Therefore, it would be desirable to have a more direct test. Now, the existence of a Fermi surface implies the presence of Friedel oscillations. For a spin liquid, this would not show up in the charge channel, but would show up in the spin channel. The challenge is how to detect this.

As is well known, oscillatory coupling has been seen between two ferromagnets separated by a paramagnetic spacer (Fig. 1) [16]. This is a consequence of Kohn anomalies of the Fermi surface which appear in the spin susceptibility [17], but are difficult to observe by neutron scattering because of their weak intensity. These anomalies are due to extremal spanning vectors of the Fermi surface. There are several types: $2k_F$ vectors, umklapp vectors, and vectors which connect different Fermi surfaces. Large vectors are difficult to observe in the oscillatory coupling since their period is comparable to the lattice constant [9] (and are therefore damped by roughness of the layers), but small vectors have been prominently observed in transition metal multilayers [16]. The existence of small vectors (long period oscillations) is generic, as large Fermi surfaces have small umklapp vectors, whereas small Fermi surfaces have small $2k_F$ vectors. Oscillatory experiments have the additional advantage of being able to detect multiple q vectors with different orientations depending on the growth direction of the layers. These oscillations are also strongly suppressed by a spin gap [18], as the latter leads to an exponential decay of the susceptibility with a distance scale set by the lattice constant [19].

We will now apply this idea to spin liquids. We start with the BEDT salt, originally discovered by Geiser *et al.* [20]. As discussed by Shimizu *et al.* [6], this material is composed of dimers that sit on a distorted triangular lattice. This particular salt, though, has a very small anisotropy of the hopping integrals of 6%. The resulting Fermi surface is shown in Fig. 2(a), which was derived from the dispersion
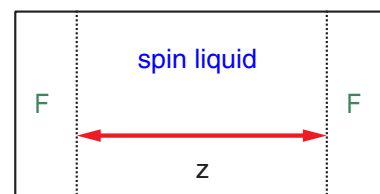

FIG. 1 (color online).   The proposed experiment involves two ferromagnetic layers (F) with a spin liquid spacer of variable thickness, z. Depending on the sign of the oscillatory coupling, the two ferromagnets will be aligned or antialigned.
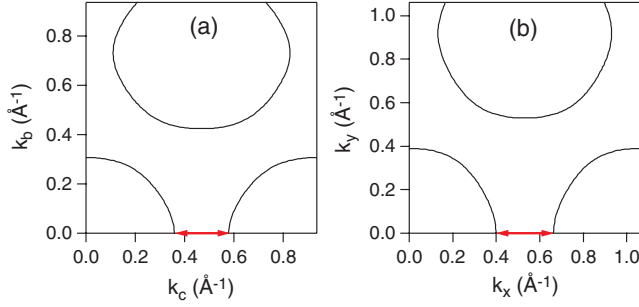
FIG. 2 (color online). Spinon Fermi surface for (a) $\kappa - (\mathrm{BEDT} - \mathrm{TTF})_2\mathrm{Cu}_2(\mathrm{CN})_3$ ($b = 8.59$ Å, $c = 13.40$ Å) and (b) $\mathrm{ZnCu}_3(\mathrm{OH})_6\mathrm{Cl}_2$ ($a = 6.84$ Å). Spanning vectors are indicated by arrows.

$\epsilon_k = 2t' \cos(k_b b) + 4t \cos(k_b b/2) \cos(k_c c/2) - \mu$ with $t = 54.5$ meV, $t' = 57.5$ meV, and $\mu = -46.2$ meV to achieve half-filling (b and c are the orthorhombic lattice constants parallel to the layers). This surface is removed by a Mott transition [21], but the transition is suppressed by pressure. We assume that the spinon surface in the Mott phase is the same as the "band" Fermi surface [22]. We note that the shortest spanning vector along k_c is an umklapp vector of length $0.94\pi/c = 0.22$ Å^{-1}, which would give rise to a real space period of 28.5 Å, comparable to that seen in multilayers [16]. We should remark that the real Fermi surface (under pressure) has been seen both by angle dependent magnetoresistance and Shubnikov–de Haas oscillations [23].

We can perform the same exercise for the herbertsmithite [7]. This material has copper atoms on a kagome lattice, with three copper atoms per unit cell. Assuming a near-neighbor spinon hopping, t_s, one has three bands, the middle of which is half-filled. Its dispersion is given by $E/t_s = 1 - X - \mu$, where $4X^2 = t_{12}^2 + t_{13}^2 + t_{23}^2 + 3t_{12}t_{13}t_{23}$ with $t_{12} = 2\cos(k_y a/2)$, $t_{13} = 2\cos(k_y a/4 + \sqrt{3}k_x a/4)$, $t_{23} = 2\cos(k_y a/4 - \sqrt{3}k_x a/4)$ and a the lattice constant (each bond has a length $a/2$). The spinon Fermi surface, previously derived by Ma and Marston [11], is shown in Fig. 2(b). It has a short umklapp spanning vector of $0.50(2\pi/\sqrt{3}a) = 0.265$ Å^{-1}, giving rise to a period of 23.7 Å, similar to that estimated for the BEDT salt.

One issue with these two materials is that they are two dimensional. To see an oscillatory period as discussed above requires that the spanning vector have a component perpendicular to the multilayer (i.e., along z in Fig. 1). This would require growing the material with a surface not perpendicular to the "(001)" direction, which may be a difficult undertaking. Along with this would be the probable difficulty of growing these materials with controlled thickness between the two ferromagnets.

We now turn to the hyperkagome case. $\mathrm{Na}_4\mathrm{Ir}_3\mathrm{O}_8$ has several advantages. It is cubic, and is also a transition metal oxide. As such, it should not have some of the growth difficulties mentioned above. This material has 12 Ir atoms

in the unit cell [8]. Each Ir atom has 4 Ir neighbors, forming a network of corner sharing triangles. Assuming again near-neighbor hopping, the resulting 12 bands can easily be found by numerical diagonalization. The spinon Fermi surface for this case has been shown by Zhou et al. [12], and consists of two hole pockets around the R point of the simple cubic zone, and one electron pocket around the Γ point [Fig. 3(a)]. The surfaces are small, and thus the shortest spanning vectors are of the $2k_F$ variety. To a first approximation, we find that the surfaces can be approximated by spheres (the cubic anisotropy is of order 10%). The k_F vector for the electron surface along (100) is $0.31\pi/a$ and that of the hole pockets $0.26\pi/a$. As there are two hole pockets, their contribution will dominate over the electron pocket in the oscillatory coupling. With a of 8.985 Å, this results in a predicted oscillatory period of 34.5 Å.

We can make a quantitative estimate of the oscillatory coupling following the literature in the GMR (giant magnetoresistance) field [17]. We assume that the coupling of the spins in the ferromagnet to those in the spin liquid can be described by an effective contact interaction $A\delta(\mathbf{r} - \mathbf{r}_i)\mathbf{s} \cdot \mathbf{S}_i$ where $\mathbf{s}$ is the spin vector of the spin liquid at position $\mathbf{r}$ and $\mathbf{S}_i$ the spin vector of the ferromagnet at position $\mathbf{r}_i$. Values for A can be estimated by calculating the transmission properties of the barrier between the ferromagnet and spacer layers, although these calculations are quite involved [18,24]. This effective contact interaction then leads to a coupling between a site in the first ferromagnet layer to one in the second

$$J_{ij} = -\frac{A^2}{16\pi^3 V} \int d^3\mathbf{q}\,\chi(\mathbf{q})e^{i\mathbf{q}\cdot(\mathbf{r}_j - \mathbf{r}_i)}, \qquad (1)$$

where V is the unit cell volume of the spin liquid, and $\chi(\mathbf{q})$ the static susceptibility of the spin liquid in units of $2\mu_B^2$ per unit cell. The interlayer coupling per unit area is then obtained by summing over the lattice sites in the second ferromagnetic layer,
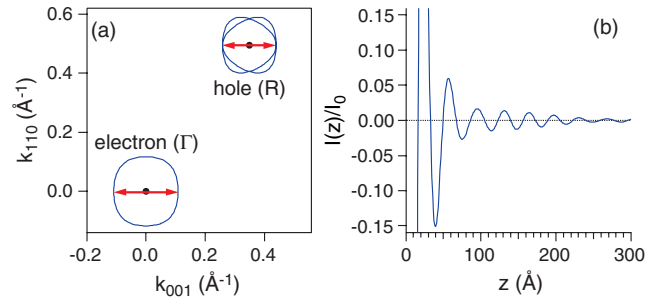
$$I_i = \frac{d}{V}S^2\sum_j J_{ij}, \qquad (2)$$

FIG. 3 (color online). (a) Spinon Fermi surface for $\mathrm{Na}_4\mathrm{Ir}_3\mathrm{O}_8$ ($a = 8.985$ Å). Spanning vectors are indicated by arrows. (b) Calculated oscillatory response from Eq. (12).

where d is the layer spacing of the spin liquid. Substituting for J_{ij}, we have

$$I(z) = -\frac{A^2 S^2 d}{16\pi^3 V^2} \int dq_z e^{iq_z z} \int d^2 \mathbf{q}_\parallel \chi(\mathbf{q}_\parallel, q_z) \sum_{\mathbf{r}_\parallel} e^{i\mathbf{q}_\parallel \cdot \mathbf{r}_\parallel} \quad (3)$$

with z oriented as in Fig. 1 and the sum is over r vectors in the second ferromagnetic layer. By Bloch's theorem, the sum reduces $\mathbf{q}_\parallel$ to zero, and we obtain

$$I(z) = -\frac{A^2 S^2 d^2}{4\pi V^3} \int dq_z e^{iq_z z} \chi(0, 0, q_z). \quad (4)$$

To proceed further, we have to specify χ. As in the spin-liquid literature [9], we will assume that χ is given by the bare polarization bubble times the Gutzwiller projection factor $g_J = 4$ [25]. This then yields

$$I(z) = -\frac{A^2 S^2 g_J g^2 s^2 d^2}{32\pi^4 V^2}$$
$$\times \int d^3 \mathbf{k} \int dq_z e^{iq_z z} \frac{f(\epsilon_\mathbf{k}) - f(\epsilon_{\mathbf{k}+\mathbf{q}})}{\epsilon_{\mathbf{k}+\mathbf{q}} - \epsilon_\mathbf{k}} \quad (5)$$

where $\mathbf{q} \equiv (0, 0, q_z)$, f is the Fermi-Dirac function, $\epsilon_\mathbf{k}$ the spinon dispersion, and g the effective g factor of the spin liquid (S is the magnitude of the spin vector of the ferromagnet, and s the magnitude of the spin vector of the spin liquid). Note that there is an implicit double sum over band indices.

The integrals over k_z and q_z can be converted to integrals over $\epsilon_\mathbf{k}$ and $\epsilon_{\mathbf{k}+\mathbf{q}}$, and the remaining integral over $\mathbf{k}_\parallel$ will be dominated by extremal q vectors connecting the spinon Fermi surface(s). The variation of the extremal q vector is given by an expansion of the spinon dispersion about k_F [26]

$$\epsilon_\mathbf{k} = v_z(k_z - k_F) + \tfrac{1}{2}(k_x^2 D_{xx} + k_y^2 D_{yy} + 2k_x k_y D_{xy}), \quad (6)$$

where v_z is the Fermi velocity along z and D is the inverse mass tensor. In our cubic case, $D_{xx} = D_{yy}$. Setting $\epsilon_\mathbf{k}$ to zero and solving, we find

$$k_z = k_F - \frac{1}{2v_z}((k_x^2 + k_y^2)D_{xx} + 2k_x k_y D_{xy}). \quad (7)$$

This is easily diagonalized by rotating 45° in k_x, k_y space

$$k_z = k_F - \frac{1}{2v_z}(k_1^2 D_1 + k_2^2 D_2), \quad (8)$$

where $D_{1,2} = D_{xx} \pm D_{xy}$.

Because of the multiple Fermi surfaces in Fig. 3(a), both intraband and interband terms are present. The latter will be particularly relevant for the susceptibility in the presence of spin-orbit coupling (known to be significant in the case of $Na_4 Ir_3 O_8$ [27]). For the intraband terms, q is simply twice k_z. Collecting these terms, we arrive at the $T = 0$ expression [17]

$$I(z) = -I_0 \frac{d^2}{z^2} \sum_n \frac{m_n^*}{m} \sin(2k_{Fn} z + \phi_n), \quad (9)$$

where m_n^* is the effective mass,

$$m_n^* = 1/\sqrt{|D_{xx}^2 - D_{xy}^2|}, \quad (10)$$

m the bare mass, and n the band index. Restoring $\hbar$, I_0 is equal to $mg_J(ASgs/(4\pi\hbar V))^2$ [28]. ϕ_n is a phase angle, equal to zero if q is a maximum, $\pi/2$ if a saddlepoint, and π if a minimum. For the interband terms, we must replace D_1 and D_2 by their averages over the two bands. For the hyperkagome case, it turns out that D_1 and D_2 are interchanged between the two hole bands. As a consequence, the interband contributions in this case reduce to the same expression as in Eq. (9), but with

$$m_n^* = 1/|D_{xx}|. \quad (11)$$

The various parameters are listed in Table I.

Note that the value of m_n^* for the interband term from Table I is 0.239. Summing over the two intraband and two interband terms, we find the total hole contribution to be twice $0.296 + 0.239$ or 1.07. The ratio of this to the electron value of 0.382 is 2.8. Noting that in the present case, for all contributions, q is a maximum [29], we then find

$$I(z) = -I_0 \frac{d^2}{z^2} (2.8 \sin(0.182z) + \sin(0.218z)), \quad (12)$$

where we have absorbed the proportionality constant for the mass (which involves the spinon hopping integral t_s) into I_0, with z (the separation of the two ferromagnets) in units of Å. This function is plotted in Fig. 3(b), where we find a periodicity consistent with the hole period of 34.5 Å mentioned above, but with beating clearly present due to the electron period. This shows the power of this technique to resolve complex Fermi surface topologies.

We note that although at very low temperatures, $Na_4 Ir_3 O_8$ exhibits a superlinear specific heat, indicating that the spinon surface might develop nodes [12], over most of the temperature range, it is consistent with an ungapped surface [13]. Therefore, we regard this cubic material as a promising one to consider in the context of our proposed experiment, though we encourage that all candidate spin liquids be looked at.

TABLE I. Properties of the hole and electron surfaces of $Na_4 Ir_3 O_8$ used in calculating the oscillatory coupling. k is in units of π/a, m^* in units of m, and ϵ in units of t_s.

Band	k_F	v_z	D_{xx}	D_{xy}	m_n^*
5	(1,1,1.260)	−0.971	−4.182	−2.465	0.296
6	(1,1,1.260)	−0.971	−4.182	2.465	0.296
7	(0,0,0.312)	1.522	2.618	−0.001	0.382

In conclusion, we have proposed an experiment based on oscillatory coupling between two ferromagnets with a spin liquid spacer for possible detection of a spinon Fermi surface. This, and other Friedel-like experiments, will hopefully be pursued in the future to see whether this novel spin liquid state exists.

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