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Detection of spinons via spin transport

Chui-Zhen Chen,[1] Qing-feng Sun,[2] Fa Wang,[2] and X. C. Xie[2]
[1]*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*
[2]*International Center for Quantum Materials and School of Physics, Peking University, Beijing 100871, China*
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The existence of deconfined spinons is the defining property of quantum spin liquids. These exotic excitations have a (fractionalized) spin quantum number and no electric charge, and have been proposed to form Fermi surfaces in the recently discovered organic spin liquid candidates. However, direct probes for them are still lacking. Here we propose to experimentally identify the spinons by measuring the spin current flowing through the spin liquid candidate materials, which would be a direct test for the existence of spin-carrying mobile excitations. By the nonequilibrium Green's function technique we evaluate the spin current through the interface between a Mott insulator and a metal under a spin bias, and find that different kinds of Mott insulators, including quantum spin liquids, can be distinguished by different relations between the spin bias and spin current. We also discuss relations to experiments and estimate experimentally relevant parameters.

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A quantum spin liquid (QSL) was first proposed by Anderson as an alternative ground state against long-range magnetic order in frustrated magnets.[1] In these systems competing spin-exchange interactions result in a large degeneracy of classical ground states, and quantum fluctuations among these states destroy long-range symmetry breaking order.[2] A particular kind of quantum spin liquid, the resonant valence bond (RVB) state, has also been proposed to be the key to high-temperature superconductivity in cuprate materials.[3,4] After decades of intense research many numerical evidences of QSL ground states have been found in semirealistic lattice models,[5–15] and many artificial parent Hamiltonians for spin liquids have been constructed.[16–18]

On the other hand, the experimental realization of spin liquids in more than one spatial dimension remains challenging until several candidate materials have been discovered recently.[2,19] Two two-dimensional (2D) triangle lattice organic salts $EtMe_3Sb[Pd(dmit)_2]_2$ and κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, the kagome lattice herbertsmithite $ZnCu_3(OH)_6Cl_2$, and a three-dimensional hyperkagome lattice $Na_4Ir_3O_8$ are found to be the most promising candidates for QSLs.[20–23] Despite structural distinctions, they are all Mott insulators with competing interactions and show no magnetic order down to temperatures that are much lower than their exchange interaction. Current measurements of magnetic susceptibility, specific heat, thermal transport, and neutron scattering have provided vital information about the properties of these materials.[24–27] However, a definitive experiment for the identification of quantum spin liquids is still missing.[2]

One of the most significant features of quantum spin liquids is that they have exotic excitations called spinons which are uncharged, and usually spin-$1/2$ mobile particles, which may obey bosonic or fermionic statistics and may or may not have a gap.[2,19] The fermionic spinons may form Fermi surfaces and are generally accompanied by an emergent gauge field.[28,29] This "spinon Fermi sea" state has received strong support from the observations of metalliclike specific heat and thermal conductivity[30] in the organic candidates at low temperatures. However, these experiments do not provide a direct proof that the mobile and possibly fermionic low energy excitations are spinons. A more reliable proof for the spinon Fermi sea would be a metalliclike spin transport in these Mott insulators.

In this Rapid Communication, we propose a four-terminal measurement of spin current through a spin liquid material (Mott insulator) as evidence for the existence of spinons. The proposed four-terminal device consists of a spin liquid material coupled to the left and right normal metal leads and each lead couples to two ferromagnetic (FM) electrodes, as shown in Fig. 1.[31] A current source is added between the two right FM electrodes. This is used to create a spin polarized current flowing from one right FM electrode through the right lead to another right FM electrode, leading to a spin-solved chemical potential (i.e., a spin bias V_R) in the right lead. Then this spin bias will drive a spin current flowing from the right lead through the spin liquid and finally into the left lead by the spinon-electron spin-exchange interaction at the interfaces between the spin liquid and the leads. Finally, a voltmeter is connected to the two left FM electrodes which are in contact with the left lead. The voltmeter is used to measure the spin bias V_L created by the spin current in the left lead. Here, the spin bias V_α ($\alpha = L,R$) is defined as the difference between the spin-↑ chemical potential $\mu_{\alpha\uparrow}$ and the spin-↓ chemical potential $\mu_{\alpha\downarrow}$ in the α lead, i.e., $V_\alpha \equiv \mu_{\alpha\uparrow} - \mu_{\alpha\downarrow}$.[32,33]

In the rest of this Rapid Communication, we will first establish the general result of the spin current through the interface between a Mott insulator in the middle region and a metallic right lead with a spin bias (see Fig. 1) by the nonequilibrium Green's function technique. We will then apply the general formalism to show that different Mott insulators can be distinguished by different relations between the spin current and the spin bias as well as temperature. We will also discuss relations to experiments and estimate experimentally relevant parameters.

The model Hamiltonian and formulation. In our theoretical analysis, we consider the model of a Mott insulator (a spin liquid or a collinear antiferromagnet) coupled to two normal leads under a spin bias on the right lead. The Hamiltonian of the system is given by $H = H_0 + H_M + H_I$, where H_0, H_M, and H_I are the Hamiltonians of the leads (metal), the middle region (Mott insulator), and the interfaces, respectively. H_0

　

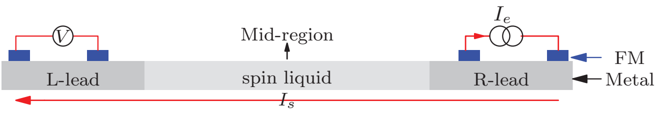

FIG. 1. (Color online) Schematic plot of a four-terminal device for the detection of spin current through a spin liquid. The four small blue bars indicate ferromagnetic electrodes. A spin polarized current is injected into the right lead (R-lead) by a current source (I_e), thus creating a spin bias and driving a spin current through the spin liquid middle region. The spin bias induced by the spin current in the left lead (L-lead) is then measured by a voltmeter (V). We will also consider other kinds of Mott insulators as the middle region instead of spin liquids.

and H_I are assumed to be

$$H_0 = \sum_{\alpha=L,R} \sum_{\boldsymbol{k},\sigma} \xi_{\alpha k\sigma} c^\dagger_{\alpha k\sigma} c_{\alpha k\sigma} \quad (1)$$

and

$$H_I = J_I \sum_{\boldsymbol{r_0}} \boldsymbol{S}_R(\boldsymbol{r_0}) \cdot \boldsymbol{S}_M(\boldsymbol{r_0}) + J_I \sum_{\boldsymbol{r_1}} \boldsymbol{S}_L(\boldsymbol{r_1}) \cdot \boldsymbol{S}_M(\boldsymbol{r_1}), \quad (2)$$

where

$$\boldsymbol{S}_\alpha(\boldsymbol{r}) = \frac{1}{2} \sum_{\mu,\mu'} \boldsymbol{\sigma}_{\mu\mu'} c^\dagger_{\alpha\mu}(\boldsymbol{r}) c_{\alpha\mu'}(\boldsymbol{r}),$$

with $\boldsymbol{S}_M(\boldsymbol{r})$ the (dimensionless) spin operator of the middle region at the position $\boldsymbol{r}$. $c_{\alpha k\sigma}(c^\dagger_{\alpha k\sigma})$ is the creation (annihilation) of the spin-σ electron in the $\alpha = $ (L,R) lead. $\xi_{\alpha k\sigma} = \varepsilon_{\alpha k} - \mu_{\alpha\sigma}$, where $\varepsilon_{\alpha \boldsymbol{k}}$ is the electron dispersion relation and $\mu_{\alpha\sigma=(\uparrow,\downarrow)}$ is the spin dependent chemical potential in the α lead. The spin-exchange interaction constants J_I are determined by the interface properties of Mott insulator and metal.[34] $\sum_{\boldsymbol{r_0}}$ means the integral over the interface. H_M depends on the type of Mott insulator we consider and will be specified later. We emphasize that there is *no* single electron tunneling term in H_I because the middle region is a Mott insulator.

Due to the spin-exchange interaction in H_I, the spin current can flow from the normal lead to Mott insulator and vice versa. When the right lead is under a spin bias, the spin current I_s flowing into the middle region from the right lead is

$$\begin{aligned} I_s &= -\left\langle \frac{d}{dt} S^z_T \right\rangle = i \left\langle \left[S^z_T, H(t) \right] \right\rangle \\ &= i\, J_I \sum_{\boldsymbol{r}_0} \left\langle \left[S^z_T, \boldsymbol{S}_R(\boldsymbol{r_0}) \cdot \boldsymbol{S}_M(\boldsymbol{r_0}) \right] \right\rangle \\ &= J_I \sum_{\boldsymbol{r}_0} \mathrm{Re}[\Gamma^<(\boldsymbol{r}_0,t,t)], \end{aligned} \quad (3)$$

with $S^z_T \equiv (\hbar/2)(N_\uparrow - N_\downarrow) = (\hbar/2) \sum_{\boldsymbol{k}} (c^\dagger_{Rk\uparrow} c_{Rk\uparrow} - c^\dagger_{Rk\downarrow} c_{Rk\downarrow})$. Here, we have used the fact that $[S^z_T, H_0] = 0$ and defined $\Gamma^<(\boldsymbol{r}_0,t,t) = i\langle S^-_M(\boldsymbol{r}_0,t) c^\dagger_{R\uparrow}(\boldsymbol{r}_0,t) c_{R\downarrow}(\boldsymbol{r}_0,t)\rangle$ with $S^-_M(\boldsymbol{r}_0) = S^x_M(\boldsymbol{r}_0) - i S^y_M(\boldsymbol{r}_0)$.

In order to solve the Keldysh Green's functions above, we first apply the equation of motion technique to solve $\Gamma^t(\boldsymbol{r}_0,\boldsymbol{k},\boldsymbol{k}',\tau,\tau') = -i\langle T_c\{S^-_M(\boldsymbol{r}_0,\tau) c_{Rk\downarrow}(\tau) c^\dagger_{Rk'\uparrow}(\tau')\}\rangle$.[35,36] By keeping the lowest order terms of J_I we have

$$\begin{aligned} \Gamma^t(\boldsymbol{r}_0,\boldsymbol{k},\boldsymbol{k}',\tau,\tau') &= \frac{-i J_I}{2N_R} \sum_{\boldsymbol{r}_0} \int d\tau_1 \chi^t(\boldsymbol{r}_0,\boldsymbol{r}'_0,\tau,\tau_1) g^t_{R\downarrow}(\boldsymbol{k},\tau,\tau_1) \\ &\quad \times g^t_{R\uparrow}(\boldsymbol{k}',\tau_1,\tau') \exp[-i(\boldsymbol{k}-\boldsymbol{k}')\cdot \boldsymbol{r}_0], \end{aligned}$$

where $\chi^t(\boldsymbol{r}_0,\boldsymbol{r}'_0,\tau,\tau_1) = -i\langle T_c[S^-_M(\boldsymbol{r}_0,\tau) S^+_M(\boldsymbol{r}'_0,\tau_1)]\rangle$ and $g^t_{R\sigma}(\boldsymbol{k}',\tau,\tau') = -i\langle T_c[c_{Rk'\sigma}(\tau) c^\dagger_{Rk'\sigma}(\tau')]\rangle$ are contour-order Green's functions for a spin operator in the middle region and free electrons in the right lead, respectively. $\Gamma^<(\boldsymbol{r}_0,\boldsymbol{k},\boldsymbol{k}',t,t') = i\langle S^-_M(\boldsymbol{r}_0,t) c^\dagger_{Rk'\uparrow}(t') c_{Rk\downarrow}(t)\rangle$ can be obtained by an analytical continuation from Γ^t, and a Fourier transform then produces $\Gamma^<(\boldsymbol{r}_0,t,t)$. Plugging the result into Eq. (3) we have

$$\begin{aligned} I_s = &\frac{J_I^2 N_\perp}{4N_R^2 N_M^2} \sum_{\boldsymbol{q},\boldsymbol{k},\boldsymbol{k}'} A_M(\boldsymbol{q},\xi_{Rk'\uparrow} - \xi_{Rk\downarrow} + V)\delta_{\boldsymbol{q}_\perp+\boldsymbol{k}_\perp-\boldsymbol{k}'_\perp} \\ &\times \{[1 + n_B(\xi_{Rk'\uparrow} - \xi_{Rk\downarrow} + V)] n_F(-\xi_{Rk\downarrow}) n_F(\xi_{Rk'\uparrow}) \\ &- n_B(\xi_{Rk'\uparrow} - \xi_{Rk\downarrow} + V) n_F(\xi_{Rk\downarrow}) n_F(-\xi_{Rk'\uparrow})\}, \end{aligned} \quad (4)$$

where N_M and N_R are the number of unit cells in the middle region and right lead, respectively. Here $N_\perp$ is the number of transverse modes (parallel to the interface) and $V = \mu_{R\uparrow} - \mu_{R\downarrow}$ is the spin bias in the right lead. $n_B(\omega)$ and $n_F(\xi_{Rk\sigma})$ are the Bose and Fermi distribution functions, respectively. $\delta_{\boldsymbol{q}_\perp+\boldsymbol{k}_\perp-\boldsymbol{k}'_\perp}$ indicates transverse (parallel to the interface) momentum conservation. The power spectrum A_M is defined as $A_M(\boldsymbol{q},\omega) = \int dt \langle S^-_M(-\boldsymbol{q},t) S^+_M(\boldsymbol{q},0)\rangle \exp(i\omega t)/[1 + n_B(\omega)]$, where $S^\pm_M(\boldsymbol{q},t)$ is the Fourier transform of $S^\pm_M(\boldsymbol{r},t) = S^x_M(\boldsymbol{r},t) \pm S^y_M(\boldsymbol{r},t)$.

We note that the behavior of the spin current is mainly determined by the power spectrum A_M of the middle region. Under appropriate conditions the momentum integrations over $\boldsymbol{q},\boldsymbol{k},\boldsymbol{k}'$ can be approximately separated, and the transverse momentum conservation factor $\delta_{\boldsymbol{q}_\perp+\boldsymbol{k}_\perp-\boldsymbol{k}'_\perp}$ will provide only a constant factor.[37] The Fermi (Bose) function will be treated exactly at zero temperature and expanded in a series of $V/(k_B T)$ at finite temperature. In the following we will apply Eq. (4) and analyze several kinds of Mott insulators as the middle region, including several spin liquids.

Spin liquids. Spinons in spin liquids may or may not be gapped. The gapped spin liquids will have exponentially vanishing spin transport at low temperature and small spin bias. Non-spin-liquid phases with a spin gap, as the valence bond crystals, will show behaviors similar to the gapped spin liquids. We therefore restrict ourselves to the types of QSLs with gapless fermionic spinons. We describe such spin liquids by the following mean-field Hamiltonian,

$$H_M = \sum_{k,\sigma} \zeta_k f^\dagger_{k\sigma} f_{k\sigma},$$

where f are fermionic spinons and $\zeta_k = \epsilon_k - \mu_s$ with μ_s the spinon chemical potential. The spinon dispersion ϵ_k may have a Fermi surface (the "spinon Fermi sea" state[28,29]) or Dirac points at the Fermi level.[38]

First, we consider the two-dimensional (2D) spinon Fermi sea case, which is the most relevant to the 2D organic spin liquid candidate materials. The spinon dispersion is

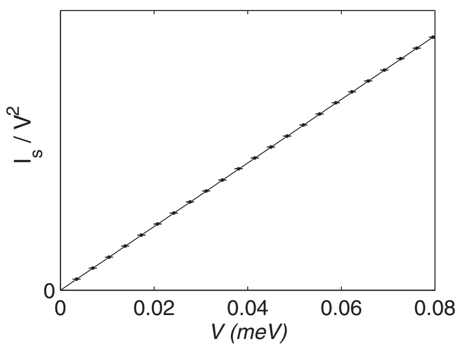

FIG. 2. In 2D spinon Fermi sea case, the scattered dots are the spin bias V dependence of I_s/V^2 at zero temperature (fitted by the simple line). The cut point at the origin directly indicates $I_s \propto V^3$.

$\epsilon_q = \hbar^2 q^2/(2m_s)$, with m_s the spinon effective mass.[39,40] The density of states of spin excitations is

$$\frac{1}{N_R^2}\sum_{\boldsymbol{q}} A_M(q,\omega) = 2\pi N_s^2\left(E_F^s\right)\omega \propto \omega.$$

Thus the spin current $I_s \propto V^3$ at $T = 0$ (see Fig. 2), and $I_s \propto (k_B T)^2 V$ at $T > 0$ with $V \ll k_B T$ (see Fig. 3).

Moreover, in the one-dimensional (1D) spinon Fermi sea case, the transverse momentum conservation factor in Eq. (4) does not exist. So the 1D case can be regarded as the special case of 2D with the transverse momentum mode number $N_\perp = 1$.

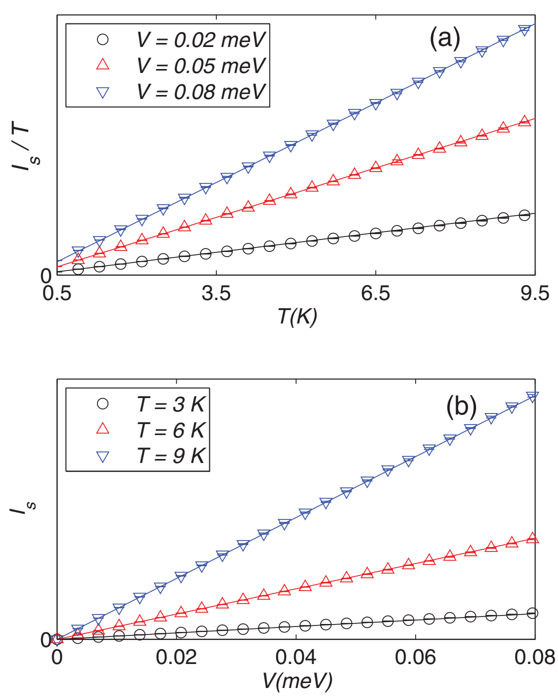

FIG. 3. (Color online) In the 2D spinon Fermi sea case, (a) the scattered dots are the temperature T dependence of I_s/T (V the spin bias on the right lead). They can be fitted by simple lines ($y \propto x$). (b) The spin current I_s vs spin bias V on the right lead at different temperatures T (fitted by simple lines). The cut point at the origin indicates $I_s \propto V$. (a) and (b) together indicate $I_s \propto T^2 V$ at high temperature $k_B T \gg V$.

In the two-dimensional Dirac spin liquid case,[38] the low energy spinon dispersion is $\epsilon_q = \pm\hbar v_F|q - q_F|$, with v_F the Fermi velocity and q_F the Fermi vector. At $T = 0$, since $\sum_{\boldsymbol{q}} A_M(q,\omega)/N_R^2 \propto \omega^3$, the spin current $I_s \propto V^5$. At $T > 0$, $V \ll k_B T$, expanding I_s in the series of $V/(k_B T)$ directly, we find $I_s \propto (k_B T)^4 V$.

Collinear antiferromagnetic Néel order. Antiferromagnetic (AFM) order is a common competitor for quantum spin liquids. Let us now consider the simplest AFM ordered state, the collinear AFM Néel order on a bipartite lattice, and show that it has a different spin transport behavior as compared to the previous spin liquid cases. We describe the spin excitations by a linearized spin wave Hamiltonian,

$$H_M = \sum_{\boldsymbol{k}} E_0\left(a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + b_{\boldsymbol{k}}^\dagger b_{\boldsymbol{k}} + \gamma_{\boldsymbol{k}} a_{\boldsymbol{k}} b_{-\boldsymbol{k}} + \gamma_{\boldsymbol{k}} a_{\boldsymbol{k}}^\dagger b_{-\boldsymbol{k}}^\dagger\right),$$

where $E_0 = 2ZS|J|$ and $\gamma_{\boldsymbol{k}} = \sum_{\boldsymbol{\delta}} \cos(\boldsymbol{k}\cdot\boldsymbol{\delta})/Z$ with δ sums over the nearest-neighbor lattice sites. Here Z is the coordination number, S is the spin quantum numbers and J is the spin-exchange constant. $b_{\boldsymbol{k}}$ ($b_{\boldsymbol{k}}^\dagger$) and $a_{\boldsymbol{k}}$ ($a_{\boldsymbol{k}}^\dagger$), are the annihilation (creation) of Holstein-Primakoff bosons[41] on the B sublattice and A sublattice, respectively.

Since the spin wave dispersion is $E_q = 4\sqrt{2}S|J|qa$, with a the lattice constant on the square lattice, the density of states of spin excitations is

$$\frac{1}{N_R^2}\sum_{\boldsymbol{q}} A_M(q,\omega) \propto \omega^2.$$

The spin current $I_s \propto V|V|^3$ and $I_s \propto (k_B T)^3 V$ at $T = 0$, and $T > 0$ with $V \ll k_B T$, respectively. All the cases we have discussed above are summarized in Table I.

Numerical estimates of the spin current. We use the following estimates of the parameters,[42] with the interface exchange coupling $J_I = 10$ meV and spin bias $V = 1\ \mathrm{K} \times k_B \approx 0.1$ meV. The metallic conductivity $\rho = 10^{-8}\ \Omega\,$m, spin diffusing length $l_s = 100$ nm, lattice constant $a = 0.3$ nm, and Fermi level $E_F^e = 1$ eV. The effective spinon mass $m_s \approx 10 m_e$ and Fermi level $E_F^s = 10$ meV. The bias induced in the left lead is evaluated by $V_L \approx (2e/\hbar)(I_s/N_\perp) * \rho * l_s$, with $N_\perp$ the number of transverse mode (see Table II).

Discussions. Many factors ignored by our analysis may affect the results. First we have assumed the conservation of the z component of spin in the entire system, so the spin current is well defined. However, in the real materials spin-orbit coupling (SOC) will generically be present.[2] We hope our results can still be applied to such systems if the linear dimensions of the sample are much smaller than the inverse of SOC. For the same

TABLE I. Behavior of the spin current through the interface between different Mott insulators (rows) and a metallic lead with respect to the spin bias V and temperature T.

	$T = 0$	$k_B T \gg V$
1D spinon Fermi sea	$\propto V^3$	$\propto (k_B T)^2 V$
2D spinon Fermi sea	$\propto V^3$	$\propto (k_B T)^2 V$
2D Dirac spin liquid	$\propto V^5$	$\propto (k_B T)^4 V$
Collinear antiferromagnet	$\propto V\|V\|^3$	$\propto (k_B T)^3 V$

TABLE II. Numerical estimates of the induced spin bias in the left lead with different kinds of Mott insulator middle regions(rows) at three different temperatures $T = 0$, 1, and 10 K. Other parameters used are given in the main text.

	$T = 0$ K (nV)	$T = 1$ K (nV)	$T = 10$ K (nV)
1D spinon Fermi sea	0.3	10	1000
2D spinon Fermi sea	0.07	3	300
Dirac spin liquid	2×10^{-7}	5×10^{-4}	3

reason we did not consider noncollinear AFM orders, e.g., the 120° order on a triangular lattice.

Second, we have ignored the emergent $U(1)$ gauge field (and the induced long-range spinon interactions) in the spinon Fermi sea and Dirac spin liquid cases. It is well known[40,43] that coupling to this gauge field can significantly change the low energy behaviors of the (spinon) Fermi sea. However, such effects have not been found in the specific heat and thermal conductivity measurements of the organic spin liquid candidates—conventional Fermi liquid behaviors were observed instead.[24,27,30] This may result from charge fluctuations which could quench the strong gauge fluctuations in the presence of the spinon Fermi sea.[44] We therefore believe our results are still valid in these materials. The effect of the $U(1)$ gauge field is an interesting theoretical question and will be left for future studies.

Finally we have assumed a clean and free spinon or magnon system in the middle region, without any scattering of spinons or magnons by interactions among themselves or impurities. We think this is not a serious problem for experiments, according to the large value of 1 μm of the experimentally estimated spinon mean free path.[30]

In summary, we have proposed to experimentally identify the spinons by measuring the spin current flowing through the spin liquid candidate materials, which would be a direct test for the existence of spin-carrying mobile excitations. By the nonequilibrium Green's function technique we evaluated the spin current through the interface between a Mott insulator and a metal under a spin bias. It was found that different kinds of Mott insulators, including quantum spin liquids, can be distinguished by different relations between the spin current spin bias as well as temperature. Although we have studied only the spinon Fermi sea and Dirac spin liquid, our general formalism can be applied to other kinds of spin liquids as well. We hope our results can stimulate more experimental studies of the spin liquid candidate materials and further promote the exchange of ideas between different fields (e.g., spintronics and strongly correlated electrons) in condensed matter physics.

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