REVIEW SUMMARY

QUANTUM MATERIALS

Quantum spin liquids

C. Broholm, R. J. Cava, S. A. Kivelson, D. G. Nocera, M. R. Norman*, T. Senthil

BACKGROUND: Years ago, Lev Landau taught us how to think about distinct phases of matter through an order parameter that characterizes the symmetry-broken state relative to the symmetry-preserving state from which it emerges. More recently, however, it has been realized that not all phases of matter are captured by this paradigm. This was spectacularly demonstrated by the discovery of fractional quantum Hall states in the 1980s. Over the years, it has been elucidated that these states, along with their exotic excitations—quasiparticles carrying a rational fraction of the elementary charge of an electron—are the consequence of topological properties of ground state wave functions with a special type of long-range quantum entanglement. One might wonder whether analogous phenomena occur for spins. Whether these "quantum spin liquids" actually exist in nature has been the subject of much investigation.

ADVANCES: Since Philip Anderson contemplated the idea of quantum spin liquids in 1973, there has been a lot of research to establish what they are and how they can be characterized. Of particular note was the realization that an effective low-energy theory inevitably resembles the gauge theory treatments also invoked in high-energy physics. However, these gauge fields are "emergent" in the sense that they reflect important structure of the many-particle state. Specifically, they describe excitations that carry a fraction of the quantum of spin in terms of emergent quasiparticles with gauge charge and/or gauge flux, analogous to the electric charge and magnetic flux in electrodynamics. One consequence is that these quasiparticle excitations can have nontrivial statistical interactions when they are braided around each other. Although most studies have focused on gapped spin liquids, equally intriguing are gapless versions—for instance, ones where the quasiparticle ("spinon") spectrum is that of relativistic electrons described by the Dirac equation. Much work has been done to address specific models and connect them to experimental analogs. This has involved a combination of analytically solvable models, as well as the development of new numerical methods that provide approximate solutions given a microscopic (lattice scale) Hamiltonian.

Perhaps most excitingly, there has been an increasingly promising effort to identify quantum spin liquids in nature. Much of the work has focused on materials where the magnetic ions reside on lattices that frustrate classical magnetic order. Examples include the triangular, kagome, hyperkagome, and pyrochlore lattices. Several candidate materials have been discovered, including organic salts, where molecular dimers realize spin-½ degrees of freedom on a distorted triangular lattice; herbertsmithite, where spin-½ copper ions form a kagome lattice; and α-$RuCl_3$, where j =1/2 ruthenium ions form a honeycomb lattice and that is thought to be proximate to the famous Kitaev model. All of these materials have properties reminiscent of spin liquids, though their documented fidelity as model systems is limited by disorder, subleading interactions, or lack of experimental information.

OUTLOOK: Given the infinite variety of potential materials and the many research groups now exploring this space, we are optimistic that a pristine materials realization of a quantum spin liquid will be discovered in the coming years. Perhaps even now a spin liquid exists in a long-forgotten drawer of a museum. Efforts to achieve ultrahigh-quality samples and new experiments designed to determine whether fractionalization and long-range entanglement occur in such materials will be key. In addition to tantalizing clues based on such techniques as thermal Hall conductivity, nuclear magnetic resonance, and inelastic neutron scattering, future methods may involve looking for spin currents to prove fractionalization, as has been done for charge degrees of freedom in the fractional quantum Hall case, or probing the range and character of quantum entanglement, as previously done in ultracold gases. Moreover, if quasiparticle excitations can be isolated and then manipulated, the prospect of a new form of topologically protected quantum computation also exists. Finally, chemically doped versions of spin liquids have been predicted to provide an unconventional route to superconductivity. The search for such phases will undoubtedly be an exciting undertaking. ■

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*Corresponding author. Email: norman@anl.gov
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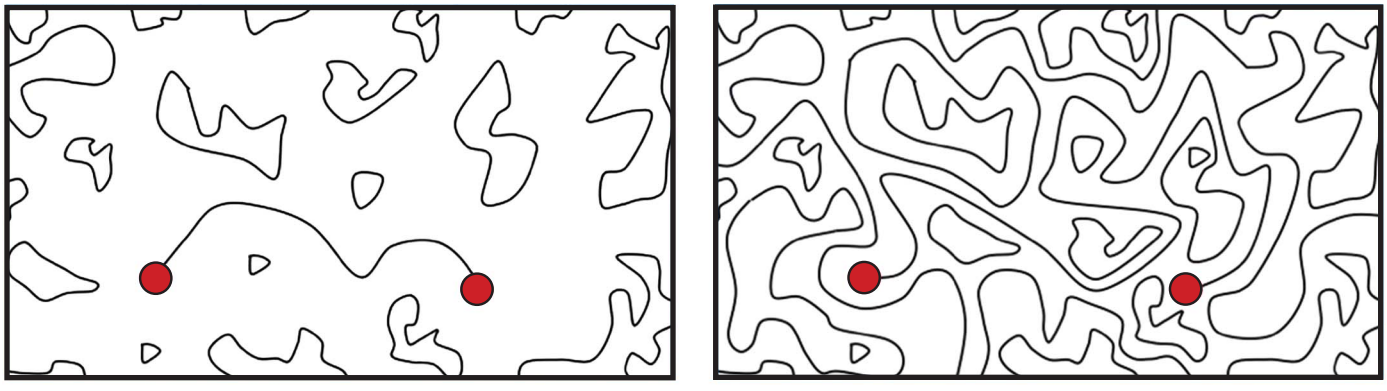

Emergent gauge theory as fluctuating loops. The loops are flux lines, with "particles" living at the ends of open lines. Left: The loops are dilute and small. The line connecting the particles costs a finite energy per unit length; the particles are confined. Right: The loops are numerous and include a fraction that are of macroscopic extent; the particles are free to move apart. This is the deconfined (spin liquid) phase.

REVIEW

QUANTUM MATERIALS

Quantum spin liquids

C. Broholm[1], R. J. Cava[2], S. A. Kivelson[3], D. G. Nocera[4], M. R. Norman[5]*, T. Senthil[6]

Spin liquids are quantum phases of matter with a variety of unusual features arising from their topological character, including "fractionalization"—elementary excitations that behave as fractions of an electron. Although there is not yet universally accepted experimental evidence that establishes that any single material has a spin liquid ground state, in the past few years a number of materials have been shown to exhibit distinctive properties that are expected of a quantum spin liquid. Here, we review theoretical and experimental progress in this area.

The history of spin liquids goes back to the early days of quantum mechanics. In 1928, Heisenberg achieved an understanding of ferromagnetism by considering a state in which all the spins point in a single direction (*1*). It is straightforward to see that a state of this sort is consistent with quantum mechanics (*2*). But problems emerged in considering antiferromagnets. Louis Néel's proposal that antiferromagnetism can be understood as a state in which the spins on alternating lattice sites point in alternating directions promoted great controversy at the time of its introduction; such a state cannot be the ground state (i.e., the lowest-energy state) of any reasonable quantum system (*3*, *4*). But it is now understood that the antiferromagnetic ground state is a prototypical example of the ubiquitous phenomenon of spontaneously broken global symmetry: The ground state is not spin-rotationally invariant and thus has a lower symmetry than the underlying Hamiltonian. This broken-symmetry point of view enables understanding of a number of universal properties of the antiferromagnetic state and their unity with similar phenomena in other ordered phases of matter. The same ideas when imported into particle physics underlie many of the successes of the standard model. For magnetic matter, it is now known that a variety of different kinds of spatially oscillating magnetic ordering patterns are possible, each corresponding to distinct broken symmetries. However, despite the successes of the broken-symmetry paradigm, the theoretical possibility of a "quantum spin liquid," for which there is no breaking of spin rotational symmetry, remained an intriguing possibility (*5*). In 1973 Philip Anderson proposed that the ground state of a simple quantum mechanical model—the spin-½ antiferromagnetic near-neighbor Heisenberg model (*6*) on a triangular lattice—might be a spin liquid. Specifically, he introduced the resonating valence bond (RVB) picture of a spin liquid wave function, based on the resonating single and double carbon-carbon bond picture developed by Linus Pauling and others to explain the electronic structure of benzene rings (*7*). Anderson's paper languished in relative obscurity until he resurrected the idea in the context of the high-temperature cuprate superconductors at the beginning of 1987 (*8*). It was realized soon afterward by Kivelson, Rokhsar, and Sethna (*9*) that the excitations of the spin liquid are topological in nature, and by Kalmeyer and Laughlin (*10*) that a version of the spin liquid could be constructed as a spin analog of the celebrated fractional quantum Hall state.

These developments in 1987 led to an explosion of interest in quantum spin liquids that continues to this day. In common with the fractional quantum Hall states, but distinct from conventional ordered states characterized by broken symmetry, the theory of the quantum spin liquid introduces new concepts, such as emergent gauge fields, into condensed-matter physics. It is not our intent here to cover the theory in great depth, as there exist several reviews (*11*–*15*). Rather, we wish to take a broader look at the field. In particular, what are the remaining big questions, both in theory and experiment?

What are quantum spin liquids?

To discuss them in the clearest context, let us focus on the idealized situation of quantum spins arranged in a periodic crystalline lattice, with interactions that are short-ranged in space. This setup describes correctly the essential physics of Mott (i.e., interaction driven) insulating materials. Mott insulating materials that do not magnetically order down to temperatures at which the spin dynamics is clearly quantum mechanical (i.e., much below the measured Curie-Weiss temperature) are attractive candidates in the search for spin liquids. However, this strategy is not sufficiently focused, as it includes nonmagnetic (quantum disordered) ground states that are not spin liquids (*16*, *17*). A more precise characterization comes from considering the structure of many-particle quantum entanglement in the ground state. A simple caricature of a magnetically ordered ground state wave function is achieved by specifying the spin on each site in the lattice. The ability to independently specify the quantum state of individual parts of a quantum many-particle system requires that the different parts have no essential quantum entanglement with each other. Thus, the prototypical ground state wave functions for conventional states of magnetic matter may be said to have short-range quantum entanglement between local degrees of freedom. By contrast, the quantum spin liquid refers to ground states in which the prototypical wave function has long-range quantum entanglement between local degrees of freedom (Fig. 1D). Under smooth deformations, such a wave function cannot be reduced to a product state wave function in real space (*18*). Such long-range quantum entanglement should be distinguished from the more familiar long-range order that characterizes broken-symmetry phases. Thus, the quantum spin liquid is a qualitatively new kind of ground state.

Just as there is no single type of magnetic order, there is no single type of quantum spin liquid. Loosely speaking, different types of quantum spin liquids correspond to different patterns of long-range entanglement. In addition, a useful (but coarse) classification distinguishes two classes of spin liquids on the basis of whether the excitation spectrum is separated from the ground state by an energy gap or not. Gapped spin liquids are simpler theoretically and are well characterized by the global topological structure of their ground state wave functions. Thus, they are said to have "topological order," a concept that also pertains to fractional quantum Hall systems. Such gapped spin liquids have well-defined emergent quasiparticles. These quasiparticles carry a topological signature that prevents them from being created in isolation (*9*, *12*). They can only be created in nontopological multiplets, which can then be pulled apart to yield multiple individual quasiparticles. A single isolated quasiparticle thus represents a nonlocal disturbance of the ground state. This nonlocality means that it can be detected far away by operations that involve moving other emergent quasiparticles. Thus, quasiparticle excitations have nonlocal "statistical" interactions (such as a charge moving around a magnetic flux). In two space dimensions, this

[1]Institute for Quantum Matter and Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, MD 21218, USA. [2]Department of Chemistry, Princeton University, Princeton, NJ 08544, USA. [3]Department of Physics, Stanford University, Stanford, CA 94305, USA. [4]Department of Chemistry and Chemical Biology, Harvard University, Cambridge, MA 02138, USA. [5]Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA. [6]Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA.
*Corresponding author. Email: norman@anl.gov

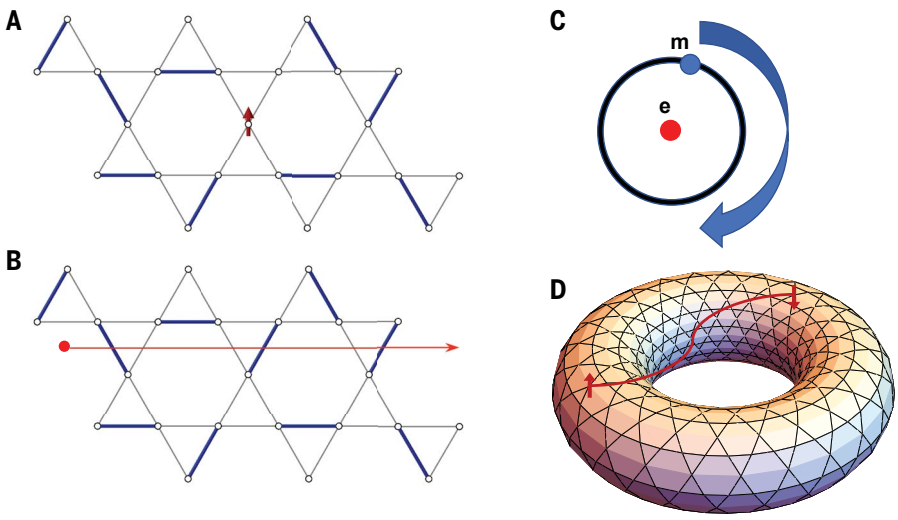

Fig. 1. Excitations of a spin liquid. Diagram of (**A**) a spinon excitation, (**B**) a vison excitation, and (**C**) braiding of anyons. Blue bonds represent spin singlets. The red arrow in (A) is a spinon, the red line with an arrow in (B) is a vison (where the phase of each singlet bond in the wave function intersected by this line changes sign), and e and m in (C) denote anyons. (**D**) Illustration of long-range entanglement of two spins, with the torus representing the ground state degeneracy typical for gapped spin liquids (the Z_2 spin liquid has a degeneracy of four on the torus associated with the topologically distinct horizontal and vertical loops that encircle the torus).

implies that the quasiparticles are "anyons" (*19*, *20*); that is, they pick up a nontrivial quantum-mechanical (Berry) phase when they circle around each other, as illustrated in Fig. 1C. This phase is associated with the "braiding" of the world lines traced by the quasiparticle trajectories.

There is a rich, formal theory of anyons in such topological ordered phases (*12*). In three space dimensions, in addition to emergent pointlike quasiparticles, there are also looplike excitations (analogous to flux lines in a superconductor) with a line tension. A quasiparticle encircling a loop excitation can also accrue a nontrivial phase. In either case (pointlike or looplike), the nonlocality associated with the quasiparticle excitation enables it to carry fractional quantum numbers associated with a global symmetry. A typical example of such a quasiparticle—known as a spinon—carries a spin of ½ and a charge of 0 (Fig. 1A). By contrast, local excitations in any insulating magnet must necessarily carry integer spin.

A second distinct class of spin liquids have a gapless excitation spectrum. In the simplest example of such a phase, the gapless spectrum admits a quasiparticle description. There also are gapless spin liquid phases where the quasiparticle description completely breaks down (*21*). In general, gapless spin liquids have power-law correlations of measurable quantities.

Given this variety of quantum spin liquid phases, what is the best theoretical framework in which we should think about them? Over the years, it has become clear that a powerful and convenient framework is provided by low-energy effective theories that involve emergent gauge fields (*22*–*25*), analogous to the vector potential in electrodynamics (*26*). Specifically, the low-energy effective theory of a quantum spin liquid is a deconfined gauge theory, that is, one in which spinons are free to propagate and thus not bound in pairs that would carry integer spin. (The particle physics analog would be a phase with free quarks.) The gauge theory description elegantly captures the nonlocal entanglement and its consequences.

To illustrate this, consider the case of a quantum spin liquid phase described by an emergent deconfined "Ising gauge field" (*27*–*30*), that is, a gauge field in which the magnetic flux can only take on two discrete values, 0 and 1. Formally, gauge theories are identified by their group structure—the Ising case is thus Z_2. Hence, this phase is known as a Z_2 quantum spin liquid. In two space dimensions, the excitations consist of a gapped excitation (the e "electric" particle) that carries Ising gauge charge and another gapped excitation (the m "magnetic" particle) that carries Ising gauge flux. These two excitations have a long-range statistical interaction: The wave function changes sign when an e particle is taken around an m particle (Fig. 1C). It is also possible to have a bound state of e and m (denoted ε). The e and m have bosonic statistics; however, their mutual braiding phase implies that ε has fermionic statistics. In systems with spin rotation symmetry, it can straightforwardly be shown that the e particle carries spin-½ (i.e., it is a spinon with bosonic statistics), whereas the m particle has a spin of 0; it is known as the "vison" (Fig. 1B). As their bound state, the ε particle also carries a spin of ½ and is known as the fermionic spinon (*31*, *32*).

There are multiple ways of thinking about how a phase with such an excitation structure might come about. A close and very useful analogy is with the excitations of the familiar Bardeen-Cooper-Schrieffer superconductor (*33*). The excitations of a superconductor include the Bogoliubov quasiparticle (resulting from the breaking up of a Cooper pair) and quantized vortices associated with $h/2e$ magnetic flux (here, h is Planck's constant and e is the electron charge). It is convenient to think about the quasiparticle in a basis where it is formally electrically neutral. In that instance, it has a braiding phase π with the $h/2e$ vortex. The Z_2 quantum spin liquid may be viewed as a phase-disordered version of a superconductor where long-range order is destroyed by quantum phase fluctuations. In this description, the fermionic spinon is identified as the cousin of the Bogoliubov quasiparticle (*26*, *34*, *35*), whereas the vison is identified as the cousin of the $h/2e$ vortex (*26*, *34*). The close relationship between the Z_2 spin liquid and the superconductor suggests that, if a spin liquid Mott insulator is found in a material, then doping it might naturally lead to superconductivity. Indeed, this is the original dream of the RVB theory as a mechanism for high-temperature superconductivity (*8*).

Other quantum spin liquid phases will have other emergent gauge groups, for example, the U(1) gauge field familiar from electromagnetism [U(1) being the group defined by rotations on a circle]; these are not obviously connected to superconductivity in any simple way. Given the importance of the gauge theory description, it is not surprising that many concepts in particle physics have been realized in the spin liquid context, including magnetic monopole-like excitations, which have been proposed in the context of the three-dimensional (3D) pyrochlore lattice (*36*). Furthermore, it is conceptually straightforward to combine features of a spin liquid with more conventional phases, giving rise to additional new quantum phases of matter with combined topological order and broken symmetries (*37*, *38*), or even new metallic phases with a Fermi surface whose enclosed volume violates Luttinger's theorem (that is, it is not proportional to the electronic density) (*39*).

Do quantum spin liquids exist in theory?

This question was settled in a variety of different ways in the late 1980s and 1990s, when the first stable effective field theory descriptions of both the Z_2 quantum spin liquid (*26*–*29*) and a different time-reversal broken version (known as a chiral spin liquid) (*40*) were developed and

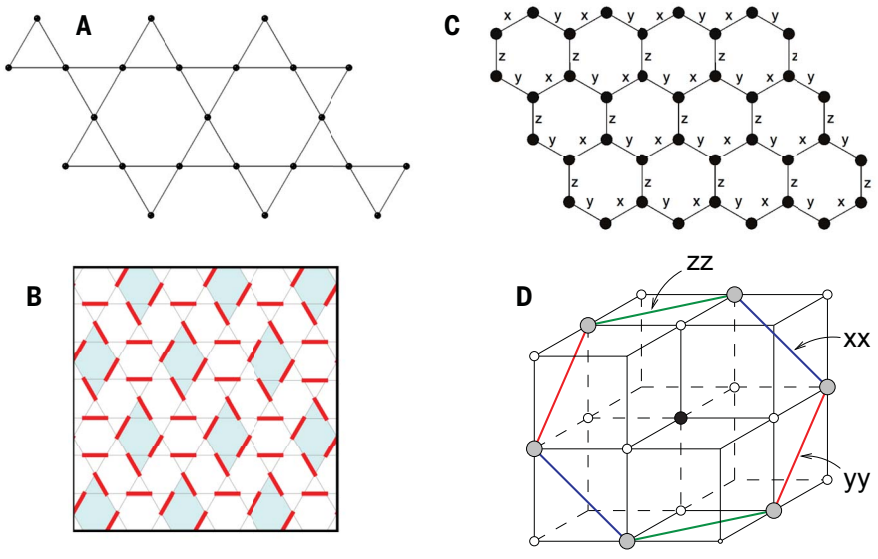

Fig. 2. Geometrically frustrated models. (**A**) Kagome lattice, (**B**) diamond valence bond solid on a kagome lattice (*153*), (**C**) Kitaev model on a honeycomb lattice, and (**D**) bond-dependent Kitaev interaction in a sixfold coordinated transition metal oxide (*67*). In (B), red bonds are singlets, with blue shading emphasizing the diamonds. In (C) and (D), x (xx), y (yy), and z (zz) denote the component of the spins involved in that bond.

their physical properties elucidated. Specific models that realize the Z_2 spin liquid were constructed in an SU(N) generalization of the SU(2) Heisenberg magnet on square lattices with short-range interactions involving more than just nearest neighbors (*28*) (so as to frustrate classical order) and on frustrated nonbipartite lattices [e.g., the triangular and kagome lattices (*41*)]. A Z_2 topological ordered state was also shown to be present in the quantum dimer model (*42*) on the triangular lattice (*43*). Additionally, Kitaev described a simple exactly solvable model (the toric code) for a Z_2 spin liquid (*44*). Building on these developments, many concrete models were constructed and reliably shown to have spin liquid phases with a variety of emergent gauge structures, in both two (*45*–*47*) and three dimensions (*46*, *48*). Though the matter of principle question has been answered in the affirmative, the question of which of these phases, if any, occur in realistic models of materials remained largely open and is still not satisfactorily settled.

Anderson's idea in 1973 that the ground state of the near-neighbor Heisenberg model was a spin liquid is not realized for the simplest form of the triangular lattice antiferromagnet, even for spin-½ systems where quantum effects are maximized, as was shown by Huse and Elser (*49*) among others. Modifications of the ideal model—for instance, the inclusion of ring exchange (*50*), further neighbor coupling (*51*), or spin anisotropy (*52*)—can, however, lead to spin liquid states (as we allude to below when talking about real materials such as the 2D organic ET and dmit salts). This led to the study of other lattices where antiferromagnetic interactions are more frustrated (i.e., act to suppress long-range magnetic order). The classic example in 2D is the lattice of corner-sharing triangles known as the kagome lattice (Fig. 2A). In the case of a near-neighbor classical Heisenberg model on a kagome lattice, continuous rotations of spins on certain clusters are possible at no energy cost (*53*–*55*), implying a large manifold of soft fluctuation modes that act to suppress order. This is particularly evident in exact diagonalization studies (*56*), which show a spectrum of states qualitatively different from the triangular lattice case, with a dense set of both singlet and triplet excitations extending to low energies. Such studies have been unable to definitively address whether the excitation spectrum for both singlets and triplets is gapped or not because of limitations of modern supercomputers [the largest lattice studied so far has been 48 sites (*57*)]. Researchers have addressed larger lattices by using approximate techniques based on quantum information–like methods, such as the density matrix renormalization group (DMRG) and various generalizations, including projected entangled pair states (PEPS) and the multiscale entanglement renormalization ansatz (MERA). The basic conclusion of such studies of the kagome lattice is that there are a number of states that have almost equal energies (*13*), including gapped Z_2 spin liquids, gapless spin liquids [so-called U(1) spin liquids where the spinons have a Dirac-like dispersion], and long-period valence bond solids. The spin liquid ground state implied by DMRG studies (*58*) appears to be a "melted" version of a 12-site valence bond solid that has a diamondlike structure, as shown in Fig. 2B, although some studies point to a U(1) gapless spin liquid instead (*59*). Exact diagonalization studies suggest that the ground state might break inversion symmetry or even be chiral in nature (*56*). Moreover, because the kagome lattice lacks a point of inversion symmetry between neighboring sites, this allows for Dzyaloshinskii-Moriya (DM) interactions that can qualitatively change the ground state relative to that of the Heisenberg model. Indeed, there are indications from simulations that the addition of DM interactions favors magnetic order (*60*–*62*).

In 2006, another exactly solvable model was reported by Kitaev (*63*). Based on a honeycomb lattice, the Hamiltonian is a less symmetric version of the Heisenberg model (*6*), where exchange on the "*x*" bonds of the honeycomb involves only S_xS_x, on the "*y*" bonds only S_yS_y, and on the "*z*" bonds only S_zS_z (Fig. 2C). Its ground state is a Z_2 spin liquid with a gapless spectrum of fermionic ε particles (known as Majoranas). Making the model anisotropic between the *x*, *y*, and *z* bonds preserves the exact solubility but gaps out the ε particle. Notably, the exact solution yields not just the ground state but the full spectrum of excitations. The manifold of states can be factored into flux sectors, with the flux referring to the product of the sign of the singlets around a hexagonal loop in the honeycomb (for the ground state, +1 for all hexagons). Flux excitations are precisely the visons mentioned above and are localized with a small energy gap. But the "unbound" Majorana is free to propagate and forms a dispersion that can be either gapped or gapless, depending on the ratio of the various *J* (J_x, J_y, J_z). The interaction of these low-energy visons with the Majoranas leads to a rather featureless spin excitation spectrum, as could be measured by neutrons (*64*). One consequence of this model is emergent fermionic statistics in the continuum of spin excitations as would be measured by Raman scattering (*65*). Even more noteworthy is the prediction of Majorana edge currents in a magnetic field, which would lead to quantization of the thermal Hall effect with a value half that expected for fermionic edge modes (*66*). Despite the seemingly contrived form of this model, it was pointed out by Jackeli and Khaliullin in 2009 (*67*) that the model might be physically realized in certain honeycomb (and "hyperhoneycomb") iridates and related materials such as α-$RuCl_3$ (Fig. 2D), which has led to an explosion of interest in both this model and those materials. This brings us to our next question.

Do quantum spin liquids really exist in nature?

Although a spin-½ antiferromagnetic chain is a 1D analog of a quantum spin liquid [and its

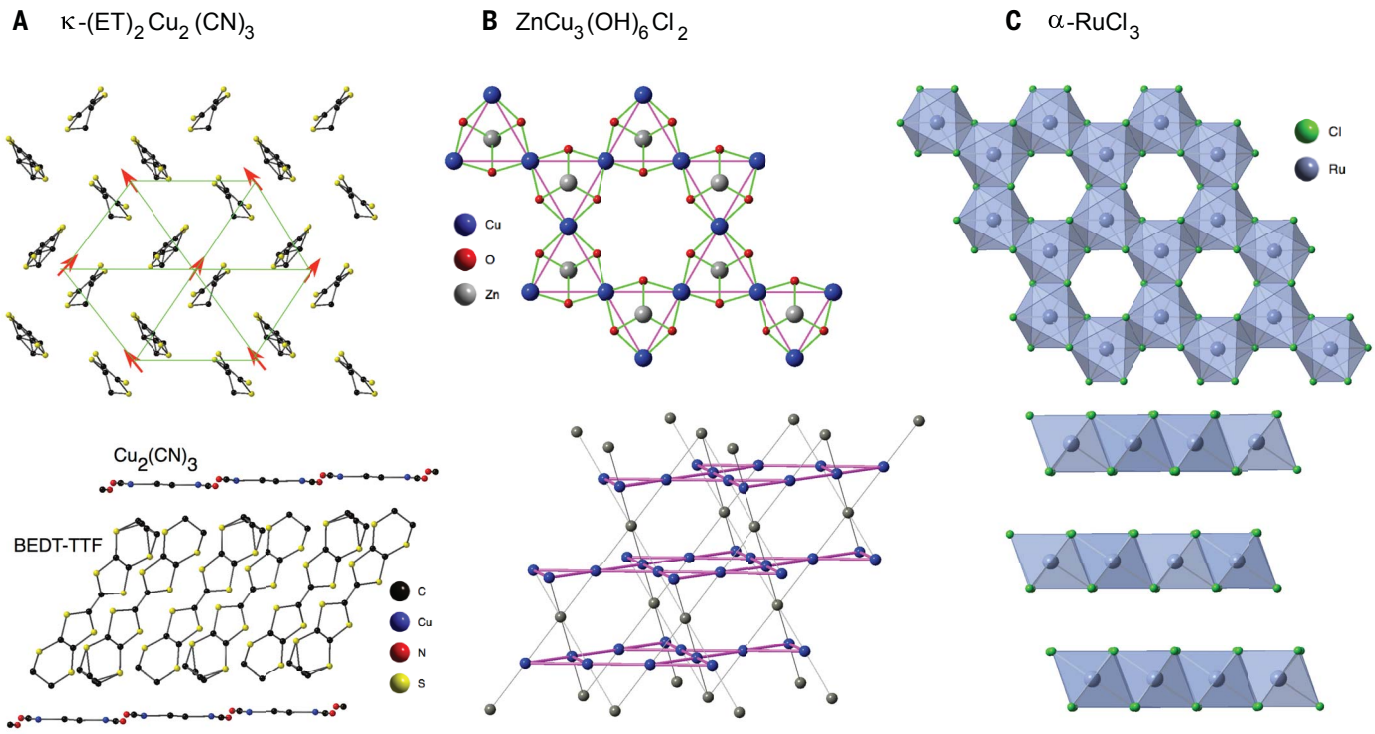

Fig. 3. Candidate spin liquid materials. Crystal structures of (**A**) κ-$(ET)_2Cu_2(CN)_3$, (**B**) herbertsmithite, and (**C**) α-$RuCl_3$. In (A), the ET dimers (top) form a triangular lattice (with the S = ½ spin degree of freedom per dimer represented by red arrows). These ET molecules are sandwiched by $Cu_2(CN)_3$ planes (bottom). In (B), Cu forms kagome layers (top) that are interconnected (bottom) by Zn (O is shown in the top only, and H and Cl have been suppressed). In (C), Ru octahedra (top) form honeycomb layers that are weakly coupled (bottom) with Cl.

spinons have been observed in experiments (*68*)], it is qualitatively different (for instance, there is no braiding in 1D). Beyond one dimension, a number of interesting candidate materials have emerged that might host quantum spin liquids, but the evidence is circumstantial. The focus has been on materials with spins on lattices that frustrate conventional Néel order. Spin-½ systems are of particular interest because they are the least classical, but the possibility of long-range entanglement for higher spin states should not be overlooked. Fluctuations are enhanced in 2D and for low coordination numbers, but even in 3D, there are pyrochlore and hyperkagome lattice systems that fail to develop magnetic order owing to geometrical frustration. Our theoretical understanding further suggests that "weak" Mott insulators that are close to the metal-insulator transition are fertile grounds for quantum spin liquid phases, consistent with the recent discovery of frustrated magnetism near the Mott transition in $(V_{1-x}Cr_x)_2O_3$ (*69*). Three of the most actively discussed classes of materials at the present time are shown in Fig. 3, and all involve lattices where either the spin, s, or the total angular momentum, j, has a value of ½. They are (i) 2D organic salts such as κ-$(ET_2)Cu_2(CN)_3$ and $EtMe_3Sb[Pd(dmit)_2]_2$, where structural dimers possessing a single spin-½ degree of freedom form a triangular lattice (*70*); (ii) herbertsmithite (and the closely related Zn-barlowite), where the Cu^{2+} ions form a kagome lattice (*71*); and (iii) α-$RuCl_3$, where the Ru^{3+} ions form a honeycomb lattice (*72*). The last two are deep in the Mott insulating phase, whereas the organic salts are weak Mott insulators close to the metal-insulator transition. We discuss each in turn, starting with the organics.

2D organic salts

Although most of these salts, where structural dimers form a (distorted) triangular lattice, have magnetic order at ambient pressure, there are a few that do not. Prominent examples are κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ (referred to here as κ-ET), κ-$(BEDT\text{-}TTF)_2Ag_2(CN)_3$, $EtMe_3Sb[Pd(dmit)_2]_2$ (referred to here as Pd-dmit), κ-$H_3(Cat\text{-}EDT\text{-}TTF)_2$, and κ-$(BEDT\text{-}TTF)_2Hg(SCN)_2Br$. Under pressure, κ-ET becomes superconducting, which was why it was first synthesized and studied (*73*). Nuclear magnetic resonance (NMR) studies show a lack of spin ordering down to temperatures well below the Curie-Weiss temperature inferred from high-temperature spin susceptibility measurements. At low temperatures, the spin susceptibility χ is a constant and the heat capacity $C = \gamma T$ has a linear temperature dependence (*74*). The Wilson ratio χ/γ is within 20% of the free Fermi gas value, which suggests that there are gapless spin-carrying excitations despite the lack of magnetic long-range order.

In Pd-dmit, despite its insulating nature, the thermal conductivity was reported to have a metallic form at low temperatures ($\kappa \propto T$) and is magnetic field dependent (*75*). If correct, this suggests that the gapless spin-carrying excitations are also mobile in this material. However, very recently this result has been reexamined in a number of dmit samples by two groups, and no such metallic thermal conductivity was found (*76*, *77*). Moreover, in κ-ET, there is at very low temperatures a dip in the thermal conductivity that, if taken at face value, might indicate a very small energy gap (*78*). This emphasizes the challenges associated with measurements of subtle features in complex materials with competing phases and the need for new results on spin liquid candidates to be thoroughly investigated. In κ-$(BEDT\text{-}TTF)_2Hg(SCN)_2Br$, heat capacity and Raman scattering indicate magnetic and electric dipole degrees of freedom that remain fluctuating to the lowest measured temperatures (*79*). Theoretically, the details of exactly which spin liquid is realized in these materials is not established. The experiments suggest that there may be a Fermi surface of emergent fermionic spinons (at least at very low temperatures). Establishing the presence of such a charge neutral Fermi surface in experiments

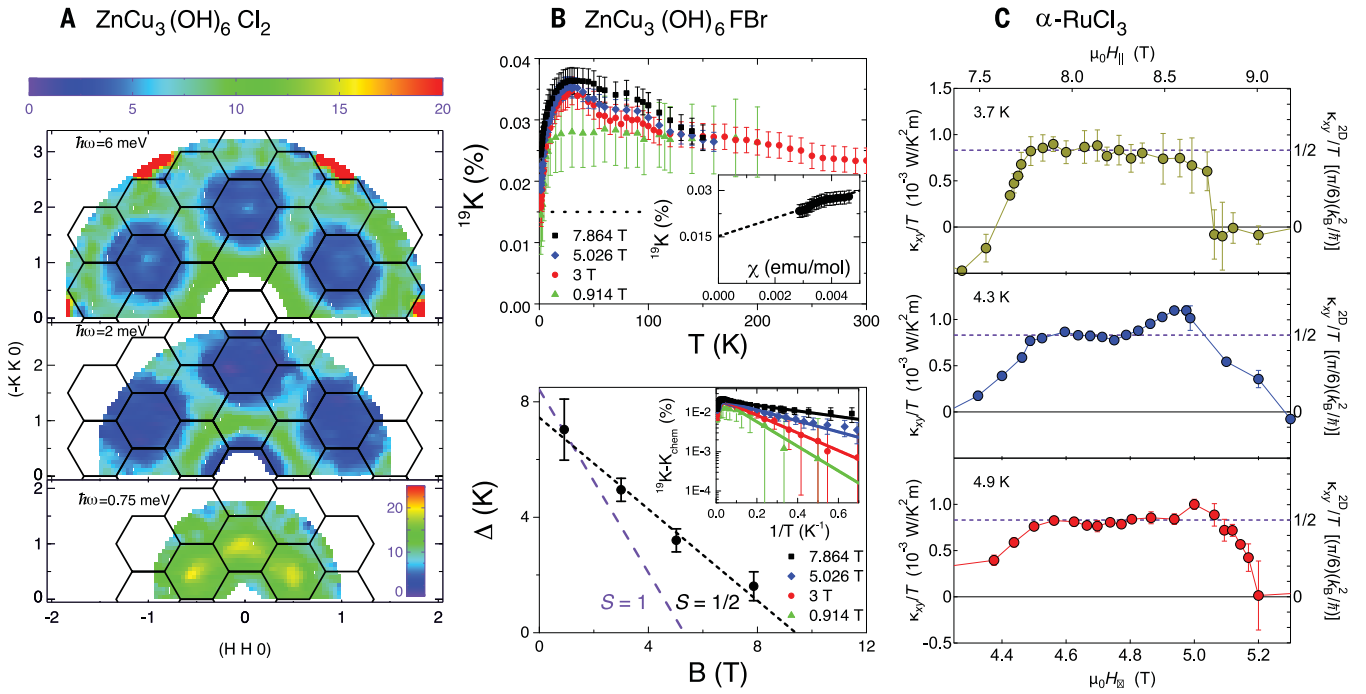

Fig. 4. Key data on spin liquid candidates. (**A**) Spin continuum of herbertsmithite from inelastic neutron scattering [S(**q**,ω) at 1.6 K in the *hk*0 plane: upper, 6 meV; middle, 2 meV; lower, 0.75 meV] (*91*). (**B**) Field dependence of the spin gap of Zn-barlowite from NMR [upper: ^{19}F Knight shift versus temperature for various magnetic fields; lower: magnetic field dependence of the spin gap, Δ, with dashed lines the expected behavior for S = ½ and S = 1 excitations] (*101*), and (**C**) quantized plateau in the thermal Hall effect of α-$RuCl_3$ [κ_{xy}/T versus magnetic field: upper, 3.7 K; middle 4.3 K; lower, 4.9 K] (*112*).

would be a great boost to our understanding. In that context, these materials (under pressure) exhibit quantum oscillations associated with their metallic Fermi surfaces. Such oscillations were found to be absent in the insulating spin liquid regime (*80*). This is contrary to the prediction that spin liquids with a spinon Fermi surface might host quantum oscillations due to weak coupling of the neutral spinons to charge fluctuations (*81*). Alternative interpretations of the data that invoke disorder to produce a heterogeneous gapless state (*82*–*84*) also deserve further experimental and theoretical exploration.

Herbertsmithite

Mineralogy has been used to inspire the search for spin liquids, making one wonder whether spin liquids are hiding in some long-forgotten desk drawer in a museum [as in the case of the first known naturally forming quasicrystal (*85*)]. The original studies (*86*) were on iron jarosites (and vanadium and chromium variants) where the magnetic ions form a perfect kagome lattice and where interesting behavior such as spin chirality has been observed (*87*). Unfortunately, these materials have long-range magnetic order, and the magnetic ions are not spin-½. However, owing to larger crystal fields and spin-orbit coupling, Ru^{3+} and Os^{3+} jarosites are candidates for a j = ½ kagome lattice, though synthesizing these minerals with 4d and 5d transition-metal ions presents an appreciable challenge.

It would be desirable to find a copper analog, given the large antiferromagnetic exchange J known to exist in copper oxides. However, the kagome ions in jarosites are 3+, and so cannot be formed with spin-½ Cu^{2+} except in diluted form. A related mineral class does contain Cu^{2+}: herbertsmithite, $ZnCu_3(OH)_6Cl_2$, a rare mineral first identified from a mine in Chile (*88*). The material was synthesized by using a hydrothermal method (*89*), and no evidence of long-range order was found. Since then, single crystals have been grown by using a refinement of the hydrothermal technique (*90*). This has allowed for single-crystal neutron scattering studies that have revealed a broad continuum of spin excitations (*91*) (Fig. 4A). Surprisingly, these excitations can be described by a dynamic magnetic correlation function of the "local" form $S(\mathbf{q},\omega) = f(\mathbf{q})\, g(\omega)$, reminiscent of the marginal Fermi liquid conjecture of Varma and colleagues (*92*). Such a form is not predicted by any known spin liquid model, though some resemblance to the data can be found in models where low-energy visons interact with the spinons, as mentioned earlier (*93*). This raises the important question of disorder. In particular, though it is claimed that the kagome spin excitations are gapped [as inferred from NMR (*94*) and neutron studies (*95*)], in reality, the entire low-energy spectrum is dominated by impurity spins (often referred to as "orphan" spins). These spins originate from the transition-metal sites between the kagome planes that are not completely inhabited by nonmagnetic Zn but also include magnetic Cu^{2+} (*96*). Similar issues exist when Zn is replaced by other 2+ ions such as Mg or Cd. Getting rid of these impurity spins is a major challenge, not only for herbertsmithite but for most spin liquid candidates where similar effects occur. This is important because some of the properties seen in herbertsmithite are reminiscent of random spin singlet states where there is a distribution of exchange energies J (*97*), and it has been claimed that the inelastic neutron scattering (INS) data can be understood in this way, as well (*98*). Such random singlet states are not quantum spin liquids (because their wave functions have a product form), even though they do exhibit quantum-critical–like scaling.

These issues have led to the study of related materials such as Zn-barlowite, which is similar to herbertsmithite except that the kagome layers are stacked differently (*99*, *100*). One advantage of Zn-barlowite is that the fluorine NMR line is simple, given its nuclear spin of ½. Analysis of these NMR data indicates a spin gap whose field dependence is consistent with a gas of spin-½ particles (i.e., spinons) (*101*) (Fig. 4B). This is further supported by INS studies, which indicate that the INS spin gap is twice that inferred by NMR (consistent

with the fact that INS measures spin-1 excitations, i.e., pairs of spinons) (*102*), though as in herbertsmithite (*93*, *94*), the low-energy properties of barlowite are dominated by defect spins. Most recently, attempts have been made to dope herbertsmithite to realize the long-sought "doped spin liquid" popularized by Anderson in 1987 (*8*). However, intercalating Li (*103*) or replacing Zn^{2+} by Ga^{3+} (*104*) leads to localized polarons [as confirmed by density functional calculations (*105*)], and thus no mobile carriers as in high-temperature superconducting cuprates (*106*). Even if polarons were not to occur, DMRG simulations predict Wigner crystallization of the doped carriers (*107*).

α-$RuCl_3$

The proposal by Jackeli and Khaliullin (*67*) that certain Mott-Hubbard systems with partially filled t_{2g}-levels and strong spin-orbit coupling might realize the Kitaev model led to an intense search. The first materials studied were those such as α-Na_2IrO_3 and α-Li_2IrO_3, where Ir^{4+} ions (with effective j = ½) form a honeycomb lattice. Although these materials exhibit long-range magnetic order, polarized resonant x-ray data show that bond-directional Kitaev interactions (Fig. 2D) indeed occur in this class of materials (*108*). This demonstrates why the recent discovery of a variant, $H_3LiIr_2O_6$, that does not exhibit long range order is important (*109*).

The realization that α-$RuCl_3$ has properties similar to those of the iridate honeycomb materials led to a huge growth in these studies. In α-$RuCl_3$, magnetic Ru is found on a honeycomb lattice between close-packed Cl planes (Fig. 3C). This material is relatively easy to grow in single-crystalline form and manipulate (as the layers are van der Waals coupled, they can be exfoliated). Also, the thermal neutron absorption cross section for Ru is a factor of 170 less than for Ir, so α-$RuCl_3$ is amenable to INS studies, which reveal a continuum of spin excitations (*110*). However, there has been some debate about which properties of this material are attributable to the Kitaev model, as opposed to more traditional physics (stemming from the non-negligible Heisenberg interaction). In particular, questions have been raised whether magnon-like excitations could explain some (or all) of the data (*111*), given that the material does order at low temperatures. Nevertheless, the spin continuum as detected in Raman data seems to obey fermionic statistics (*65*). Most notably, magnetic order is suppressed upon applying a magnetic field, implying that a spin liquid phase might exist in a range of magnetic fields. This led to a measurement of a thermal Hall signal that plateaued in a small range of temperature and magnetic field (*112*) (Fig. 4C). The value of this plateau is consistent with Majorana edge modes, being one-half of the value for fermionic edge modes (*66*). The observation of such a quantized plateau is peculiar, given that the thermal Hall angle is small (the longitudinal thermal conductivity is dominated by phonons), but this has been explained by two different theory efforts (*113*, *114*). As with most important experiments in this field, this result has yet to be reproduced by other groups. In addition, consistent with the organics and herbertsmithite, disorder should play an important role as well, particularly given the presence of stacking faults (*115*). Finally, based on the evidence that α-$RuCl_3$ exhibits spin liquid behavior, it is of great interest to study the physical properties of electron- and hole-doped variants (*116*, *117*).

A big question looms for the honeycomb-based spin liquid candidates: Is the Kitaev model actually relevant to these materials? The spin liquid in the exact solution may have only a tiny regime of stability beyond the solvable limit, on the basis of numerical calculations of the Kitaev model supplemented with Heisenberg exchange interactions (*118*). Furthermore, in the exact solution, the vison gap is very small (only a few percent of the Kitaev exchange) and so thermally, the spin liquid state only occurs at very low temperatures. Recent calculations suggest that a certain spin-anisotropic "symmetric exchange" enhances the stability of the exactly solved spin liquid (*119*). Alternatively, the possibility that any spin liquid that occurs in α-$RuCl_3$ or the iridates may not be smoothly connected to the Kitaev spin liquid must be kept in mind (*120*).

Other candidate materials

Space considerations preclude a detailed account of other spin liquid candidates. Of recent interest has been $YbMgGaO_4$, where the Yb ions form a triangular lattice, albeit with disorder on the nonmagnetic cation site. It is easy to grow and study, and the small energy scales associated with the 4f Yb ion make it more amenable to certain types of studies [extensive neutron scattering studies have been done (*121*)]. It, too, has been claimed to possibly have a "spinon" Fermi surface (*122*), but as with most spin liquid candidates, disorder plays an important role (*82*, *123*)—in this case, Mg and Ga interchanges that distort the Yb environment (*124*). Similar considerations apply to $Ba_3CuSb_2O_9$ (*125*), where Cu/Sb interchanges occur. Another candidate, $Ca_{10}Cr_7O_{28}$, can be described as a triangular lattice of six Cr^{5+}-based spin-½ clusters—each consisting of an antiferromagnetic and a ferromagnetic triangle interacting ferromagnetically with each other. Extensive experimental and numerical work on this bilayer kagome material has established its spin Hamiltonian and the lack of static spin ordering at temperatures as low as 0.3 K (*126*). For both $YbMaGaO_4$ (*127*) and $Ca_{10}Cr_7O_{28}$ (*128*), however, the absence of a linear term in the thermal conductivity argues against the existence of a spinon Fermi surface. Moreover, a lack of long-range magnetic order has been reported in the triangular-based materials $NiGa_2S_4$ (*129*), $Ba_8CoNb_6O_{24}$ (*130*), $NaYbO_2$ (*131*), and $Ba_4NbIr_3O_{12}$ (*132*), as well as in the honeycomb-based material $BaCo_2As_2O_8$ (*133*). Recently, a copper oxide, averievite [$Cu_5V_2O_{10}$(CsCl)], was identified in which the copper ions form a pyrochlore slab. First discovered in a volcano in Kamchatka, the material was synthesized and subsequently languished in an academic thesis, only to be "rediscovered" (thanks to Google Scholar) (*134*). Substitution by zinc likely replaces the intersite copper ions (as in herbertsmithite), isolating the copper kagome layers, and the resulting susceptibility and specific heat are reminiscent of herbertsmithite (*132*). Several materials are also known where magnetic ions form a "hyperkagome" lattice (obtained by taking the kagome layer and pulling it into the third dimension). Of particular note are $Na_4Ir_3O_8$ (*135*) and $PbCuTe_2O_6$ (*136*), but again both have quenched disorder (for the former material, caused by partial occupation of the Na sites) and distortions (for the latter material, there are many exchange parameters associated with its distorted hyperkagome lattice). As for other frustrated 3D lattices, extensive studies on rare-earth and transition-metal pyrochlores are beyond the scope of this article, and the reader is referred to a recent review (*137*). An exciting recent proposal (*138*), which has received some experimental support (*139*, *140*), is that the layered transition-metal dichalcogenide 1T-TaS_2 might be a quantum spin liquid.

The future

This review of quantum spin liquids may leave one to ask, "What else is out there?" Almost certainly, a lot. As for materials, many interesting ones known in mineralogical form have yet to be made in the lab and studied for their magnetic properties. As an example, quetzalcoatlite (named after an Aztec god) has copper ions on a perfect kagome lattice (*141*). But it, like many other minerals, is known only by its structure and nothing else. A systematic study of potentially frustrated magnetism in mineral collections might be a good start, followed by attempts to make cleaner synthetic versions of the most-promising minerals. A recurrent challenge with frustrated magnets is that chemical disorder acts at the "ultraviolet" scale, giving rise to orphan spins. Clearly, more attention (and resources) needs to be devoted to synthesis, both in developing promising new synthesis routes (high pressure, hydrothermal, molecular beam epitaxy, etc.) and finding ways to mitigate and control disorder. This is a difficult task, but it is useful to recall that it took decades

of materials research to develop elemental silicon clean enough for applications and modulation-doped AlGaAs heterostructures that display the fractional quantum Hall effect. Disorder—especially when carefully controlled—can also be illuminating. The tantalizing possibility of replacing Fe by Ru or Os in jarosites has been mentioned above. Similarly, one wonders what the osmium analog of α-$RuCl_3$ would be like (*142*). And, of course, obtaining a doped spin liquid that is metallic would be the holy grail for many (*8*, *9*, *22*, *143*). This could potentially be accomplished by ionic liquid gating to avoid chemical disorder.

Having addressed materials-based issues above, we turn to theory. Although great strides have been made in numerical techniques, we still do not know, for instance, what the ground state is of the near-neighbor Heisenberg model on a kagome lattice, and less about many other frustrated lattices, or for "real" Hamiltonians that contain multiple exchange parameters as well as anisotropic exchange and DM terms. Still less is known about dynamical and non-equilibrium properties. Although neutron scattering when combined with theoretical calculations of the magnetic structure factor $S(\mathbf{q},\omega)$ can provide circumstantial evidence for a spin liquid (*144*), methods to probe entanglement are needed to obtain model-independent evidence. As spin liquids are spin relatives of the fractional quantum Hall effect, it would make sense to apply methods known from spintronics to search for spin currents (*145*, *146*), the spin Hall effect, spin noise, and other spin-related effects that might expose the nature of the spinons (if they indeed exist). As for visons, a proposal for their study was made many years ago (*147*) that involves looking for trapped magnetic flux in a spin-liquid ring. This experiment was actually performed on a superconducting cuprate with a null result (*148*), but obviously doing this sort of experiment on spin liquid candidate materials would be in order. Similarly, impurities can be exploited not only to trap Majorana fermions but also to induce Friedel oscillations near defects (that could be detected by a scanning tunneling probe) that could reveal a spinon Fermi surface should it exist (*149*). And tunneling has been advocated as a possible probe of how electrons could potentially fractionalize when injected into a spin liquid (*150*). Ultimately, if topological excitations were identified in a material, then one could think about probing and extending their phase coherence time and braiding them in steps toward their utilization for "topological" quantum computation (*151*) (Fig. 1C). As for other potential applications, we can think of no better way to end than with Michael Faraday's supposed response to William Gladstone's dismissal of a scientific discovery: "What use is it?" he quipped. "Why, sir, there is every probability that you will soon be able to tax it."

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Quantum spin liquids

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An overview of an exotic type of liquid

Materials with interacting quantum spins that nevertheless do not order magnetically down to the lowest temperatures are candidates for a materials class called quantum spin liquids (QSLs). QSLs are characterized by long-range quantum entanglement and are tricky to study theoretically; an even more difficult task is to experimentally prove that a material is a QSL. Broholm *et al.* take a broad view of the state of the field and comment on the upcoming challenges.

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