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Do quantum spin liquids exist?

Takashi Imai
and Young S. Lee

The search for the hypothetical state has been a 43-year-long slog, one whose end may now be in sight.

It was 1987, a year after high-temperature superconductivity was discovered in the cuprates. Over a lunch table in a small Chinese restaurant in downtown Tokyo, a group of physicists were excitedly discussing Philip Anderson's new paper,[1] which proposed that the insulating phase of the cuprates is a quantum spin liquid (QSL). Similar conversations probably took place among physicists around the world.

One of us (Imai), then a young graduate student, wondered aloud what the big deal was. An unamused old-timer snapped, "You can surely try to read the new paper!" Still, he went on to explain that in 1973 Anderson had examined the possibility of a peculiar destruction of magnetism exhibited by spins arranged in a triangular lattice.[2] The new paper extended his original work to the square-lattice geometry found in the newly discovered cuprate superconductors.

The QSL, in theory, represents a new state of matter. Unlike conventional magnetic states, such as the ferromagnetic state with parallel spins (figure 1a) or the antiferromagnetic Néel state with antiparallel spins (figure 1b), a QSL never enters into a long-range ordered phase with a static arrangement of spins. Instead, the electrons' spins remain fluid-like, even at absolute zero temperature. Due to quantum effects, the spins perpetually fluctuate without breaking symmetry.

The discovery of cuprate superconductors had broader physics implications than just raising the superconducting transition temperature above the boiling point of liquid nitrogen. Lanthanum cuprate (La_2CuO_4) and related materials turn out to be ideal platforms for exploring the quantum effects of magnetism in low-dimensional systems.

The spin-½ Cu^{2+} ions in La_2CuO_4 are arranged neatly in a square lattice. Reducing the spin-projection quantum number S_z of the spin-up state by 1 results in the spin-down state and reverses the orientation of the magnetic moment. Therefore, quantum effects manifest themselves spectacularly for magnetic materials with spin $S = ½$.

The trouble with antiferromagnetism

The fundamental ideas behind the QSL state trace back to a related debate from decades earlier over the concept of antiferromagnetism.[3] To examine the issues at hand, first recall what ferromagnets are. Above the Curie temperature T_C, thermal fluctuations with an energy scale k_BT, where k_B is Boltzmann's constant and T is temperature, overwhelm the spin–spin exchange energy J. Hence the spins are correlated only over short distances and remain dynamic. But when the ferromagnet is cooled below T_C, the spins undergo a phase transition and point in a unique orientation that minimizes the total energy.

The underlying physics of ferromagnetism may be described by the Heisenberg Hamiltonian,

$$\hat{H} = J\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j,$$

where $\mathbf{S}_i$ represents the spin operator of the ith atom and the summation is taken over nearest neighbor pairs on the lattice. If J is negative, as is the case for ferromagnets, parallel spins minimize the total energy to about $-|JS^2|$ per spin. In other words, the ground state of the ferromagnetic Heisenberg model is an eigenstate with parallel spins, $|\uparrow\uparrow\uparrow\ldots\rangle$.

In contrast, the case of an antiferromagnetic Heisenberg model with a positive J between nearest neighbors is not trivial.[3] Classically, a Néel state—named for Louis Néel, who predicted

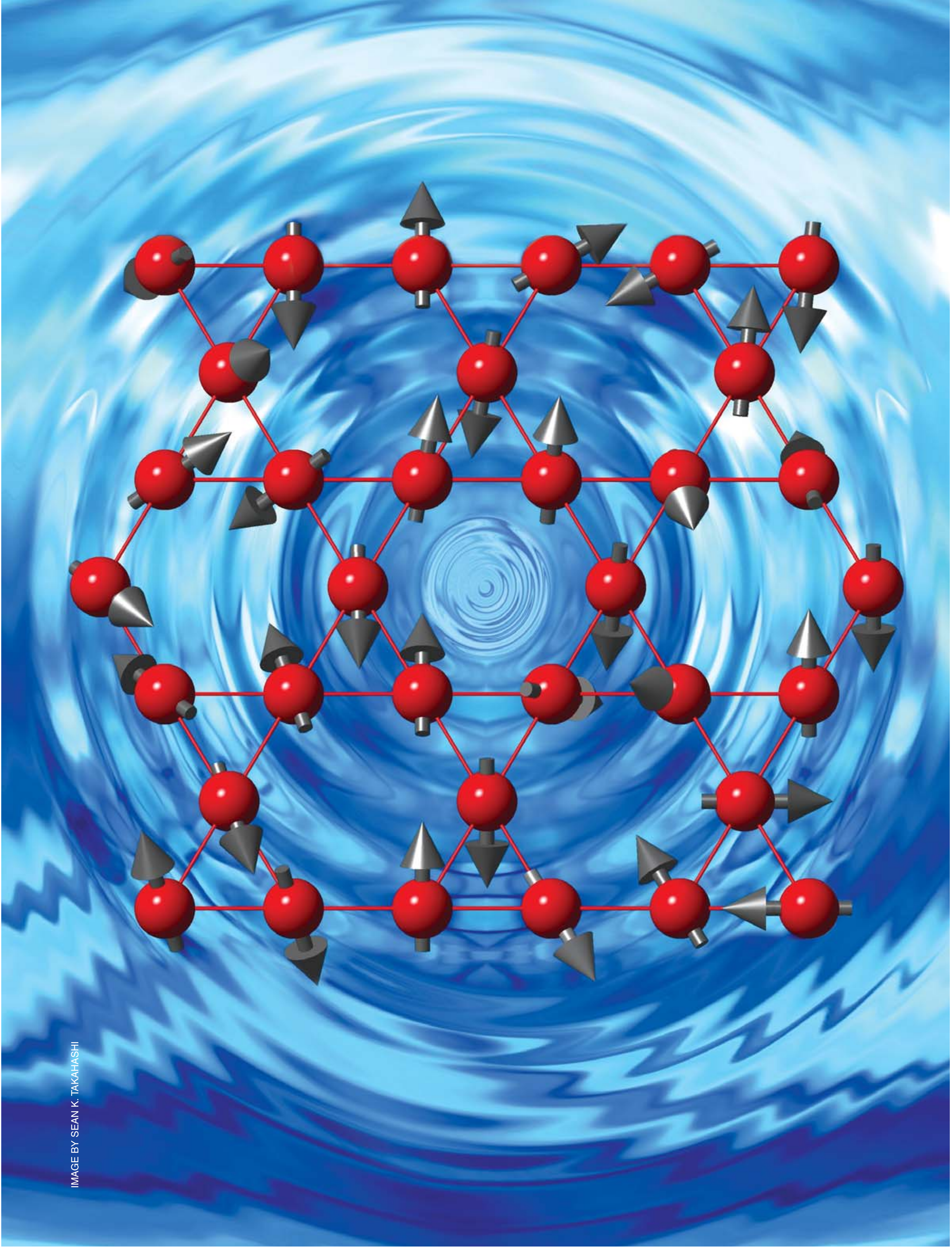


IMAGE BY SEAN K. TAKAHASHI

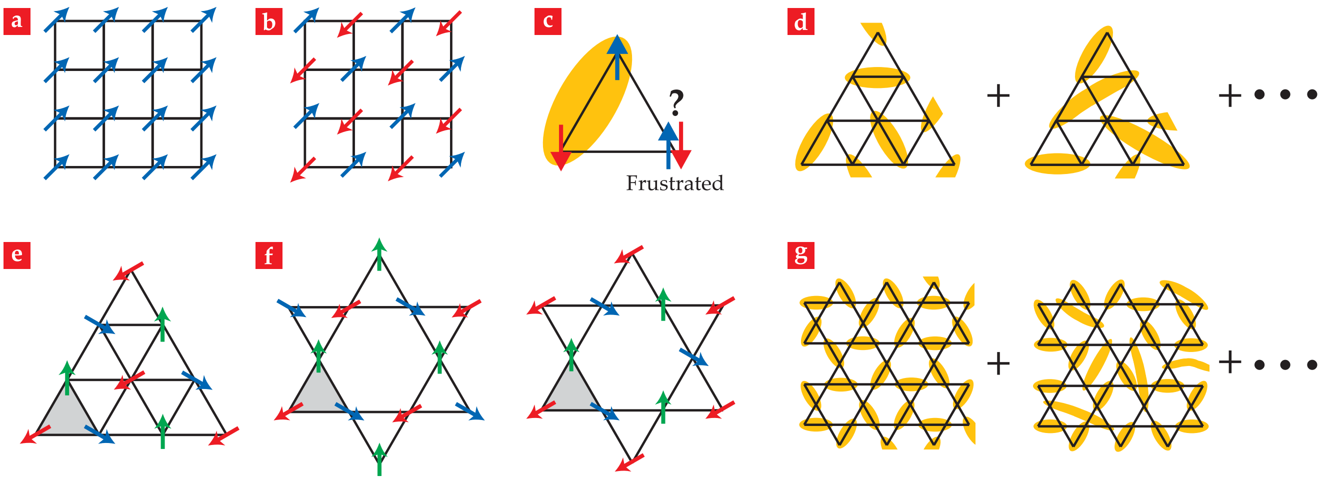


FIGURE 1. FRUSTRATION EFFECTS. (a) Ferromagnetic and **(b)** antiferromagnetic ordered states can exist on a square lattice without frustration. The arrows represent spins, and their colors identify their orientations. **(c)** Three $S = ½$ spins arranged at the corners of an equilateral triangle have more difficulty. When two spins form a singlet state with an antiparallel configuration (represented by the oval) to minimize the energy, the third spin cannot decide which direction to point. **(d)** On the edge-sharing triangular lattice, the resonating valence bond state is a linear superposition of various singlet patterns. Ovals represent two spins forming a singlet pair. **(e)** Classically, the 120-degree order—a compromise between antiferromagnetic interactions and geometric constraints—minimizes the energy. Once the spin configuration is set for one triangle—the shaded one at the lower left corner, say—the spin configuration is set throughout the entire lattice. **(f)** The 120-degree compromise works less well on a kagome lattice with corner-sharing triangles. Setting the spin configuration of the lower left triangle does not constrain that in the rest of the lattice. The spin configurations shown here are two among the numerous degenerate states that classically have the same lowest energy. The indecisiveness of spins to settle with one particular configuration favors a quantum spin liquid. **(g)** A quantum spin liquid state in the kagome lattice is a linear superposition of the collective singlet states.

its existence—with alternating spin orientations on neighboring sites, $|\uparrow\downarrow\uparrow\downarrow\ldots\rangle$, would minimize the total energy to about $-|JS^2|$ per spin as well. Quantum mechanics, however, prohibits such a stationary state for the Heisenberg Hamiltonian. A basic result in quantum mechanics for a system of two $S = ½$ spins is that neither the $|\uparrow\downarrow\rangle$ nor the $|\downarrow\uparrow\rangle$ state is an eigenstate of the two-spin operator $J\mathbf{S}_1 \cdot \mathbf{S}_2$. A linearly superposed singlet state, $(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)/\sqrt{2}$, is the ground state, with a total spin of zero for $J > 0$.

Lev Landau therefore suggested that the Néel state $|\uparrow\downarrow\uparrow\downarrow\ldots\rangle$ would not exist. Instead, as a result of quantum fluctuations, spins would form a many-body analogue of the spin-singlet state with superposed up and down spins at each site. Even today, exact analytic results for $S = ½$ antiferromagnetic Heisenberg models are generally not known.

It took the invention of neutron diffraction during the post–World War II era to experimentally establish the existence of the Néel state. Since then, it has been found in a wide range of antiferromagnets on three-dimensional lattices. In the actual Néel state, the magnitude of the ordered magnetic moment is less than $\sqrt{S(S+1)}$ Bohr magnetons (the magnetic moment of a single free electron is about 1 Bohr magneton), an indication that quantum mechanics is indeed at work. In accordance with Landau's arguments, zero-point fluctuations have the effect of shrinking the time-averaged magnetic moment. In addition, the elementary excitations, called magnons, that arise from the Néel state have been measured by inelastic neutron scattering experiments.

The Néel state strikes back

In his classic 1973 paper,[2] Anderson proposed that in an edge-sharing triangular lattice, geometrical frustration could prevent spins from undergoing magnetic long-range order into a Néel state. The reasoning is that three spins at the corners of a triangle cannot be made mutually antiparallel (figure 1c). Instead, quantum fluctuations could stabilize a so-called resonating valence bond (RVB) state.

In the RVB picture of chemical bonds in aromatic molecules such as benzene, the carbon–carbon bonds may be envisioned as a fluid of mobile single (C–C) and double (C=C) valence bonds that dynamically alternate their positions. In analogy, Anderson posited that neighboring $S = ½$ spins in a triangular lattice would form singlet pairs, but the spins would constantly alter their singlet partners and rearrange the pairings (figure 1d). The resulting RVB state is a liquid-like state of spins—an example of a QSL.

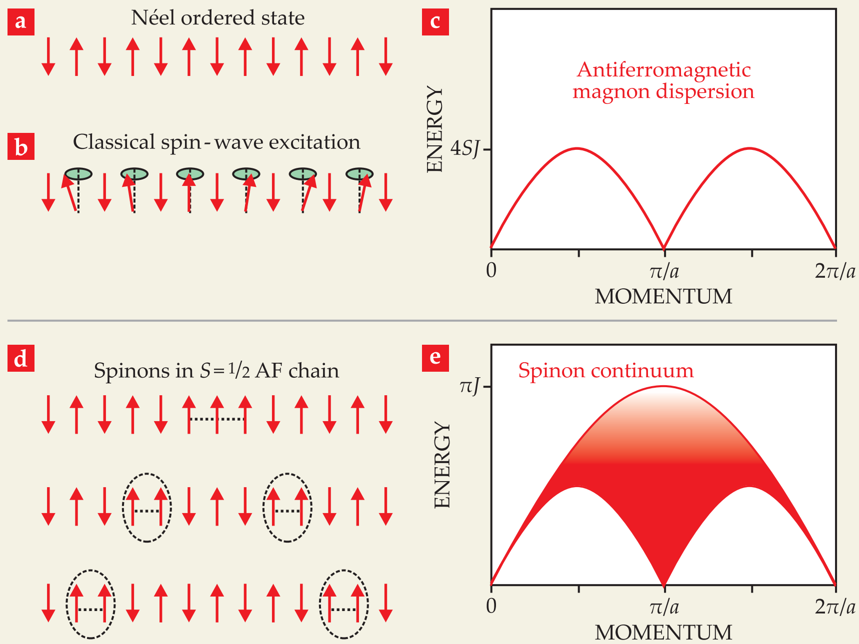
It turns out, however, that even edge-sharing triangular-lattice Heisenberg antiferromagnets can still form a Néel ordered state. In the so-called 120-degree structure (figure 1e), the spins compromise by pointing at 120° relative angles.[4]

In 1987 Anderson infused new life into the QSL idea by studying it in the context of antiferromagnetically interacting $S = ½$ Cu^{2+} spins in La_2CuO_4, the undoped parent material of the newly discovered cuprate superconductors.[1] But once again, subsequent experimental and numerical research confirmed that the conventional Néel ordered state is the ground state[5] even for $S = ½$. Thus, time and again, the ubiquitous Néel state appeared to be favored over the QSL ground state.

Unusual suspects

Nonetheless, the search for new candidate materials that may harbor a QSL continued.[6] Over the past decade, two classes of materials have emerged as promising avenues to realize a

SPIN WAVES VERSUS SPINONS



When the magnetic moments of a magnet enter an ordered state, the low-energy excited states are known as spin waves and the quantum of excitation is called a magnon. How the spin waves behave reveals the symmetry and strength of interactions and other fundamental properties of the magnetic system. In the high-temperature paramagnetic phase, the moments are equally likely to point in any direction. But in the low-temperature ordered phase, the moments must spontaneously select a unique direction along which to align the macroscopic magnetization. Goldstone's theorem says that a consequence of that spontaneous breaking of a continuous symmetry is that the energy of the longest-wavelength spin waves will vanish.

Consider the simple example of antiferromagnetically interacting magnetic moments arranged on the one-dimensional lattice shown in panel a of the figure. The classical ground state has neighboring moments that are antiparallel. The Néel ordered state consists of two interpenetrating sublattices, an up sublattice and a down sublattice. Panel b shows schematically a spin wave through which the reduction in magnetization of the up sublattice is shared by all the moments on that sublattice—for simplicity, the perturbation of the down sublattice is omitted. The energy of the spin wave depends on its wavelength, and the associated magnon energy–momentum dispersion relation is shown in panel c. The magnon with wavevector π/a (the antiferromagnetic ordering wavevector for a crystal with lattice parameter a) is the Goldstone mode with zero energy. The magnons with other wavevectors are well-defined modes, which disperse to energies up to $4SJ$, where J is the spin–spin exchange energy.

For quantum spins with $S = ½$ arranged on a 1D antiferromagnetic (AF) chain, the competition with energetically favorable singlet states results in large quantum fluctuations out of the ordered state. Hans Bethe in 1931 showed that the ground state is not Néel ordered. Antiferromagnetic spin correlations exist, but they are short ranged. The 1D $S = ½$ antiferromagnetic chain is the first known example of a quantum spin liquid.

Because spin-rotational symmetry is not broken, the low-energy excitations cannot be classical spin waves. Rather, the $S = 1$ quantum of excitation is carried by two $S = ½$ spinons. An illustration of spinon excitations on an $S = ½$ 1D AF chain is shown in panel d. A single flipped spin results in two unsatisfied bonds (dashed lines). Those unsatisfied bonds can then freely propagate along the lattice and act as domain boundaries between short-ranged antiferromagnetic regions. Due to the larger phase space for satisfying momentum and energy conservation, the allowed energies for the spinons (dashed ovals) form a continuum, indicated by the red shaded region in panel e. This is in marked contrast to the well-defined modes of magnon excitations.

One may think of the $S = 1$ magnon excitation as having fractionalized into two $S = ½$ spinons. Spinons, therefore, contribute to bulk properties, such as specific heat, in ways that are distinct from magnons. Also, note the difference in periodicity along the momentum axis for the dispersions in panels c and e. Because the quantum spin liquid does not have long-range antiferromagnetic order, it retains the same periodicity associated with the structural unit cell.

QSL in two dimensions. One is the kagome Heisenberg antiferromagnet,[7] named after the Japanese word for a woven-basket pattern consisting of a corner-sharing triangular lattice (figures 1f–g). The other is quasi-2D molecular solids with an edge-sharing triangular lattice structure (figure 2a), which are on the verge of a metal–insulator transition.[8]

Direct experimental proof of a QSL state is hard to come by. One of the unique features of a QSL is that the ground state does not follow the conventional Landau paradigm for phase transitions. Typically, interactions between particles in a collection will lead to ordering at low temperatures, and will thereby lower the overall symmetry. For example, the ferromagnetic transition breaks spin-rotational symmetry because the static moment of the electrons or ions must point in a specific direction. A local order parameter exists and becomes nonzero below a transition temperature. As a result, measurements of heat capacity, magnetization, and other thermodynamic quantities as a function of temperature show sharp anomalies in the vicinity of the transition temperature.

For a QSL, a local order parameter does not exist. The spins do not order and hence do not break spin-rotational symmetry, nor do they form singlets that are fixed to the crystal lattice and hence do not break translational symmetry. No phase boundary is crossed on cooling. So initial experimental tests for the QSL state in a material consist of looking for the absence of a phase transition. Measurements showing that the spins interact strongly yet fail to order, even at temperatures well below the interaction energy scale, would be consistent with a QSL state.

The question that immediately follows is whether the lack of ordering is a consequence of defects in the system. It is well known that quenched disorder in a spin system can lead to spin-glass physics, in which the dynamics slow down dramatically or freeze below a glass temperature without having long-range order.[9] Such spin freezing can be seen in magnetization measurements as a divergence between the temperature dependences of the spin susceptibilities of samples cooled in zero and nonzero magnetic fields. The absence of a glass transition at a temperature scale well below the interaction-energy scale would count as further evidence, albeit indirect, of the QSL state.

Terms of entanglement

The triangular-lattice molecular solids, mentioned earlier, pass all initial tests for being QSLs.[8] Specifically, they are κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$. Here, BEDT-TTF and dmit are particular organic molecules and Et and Me are ethyl and methyl groups. The structure of EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ is depicted in figure 2a. In the materials, the interaction energy scale J/k_B, deduced from the temperature dependence of the spin susceptibility, is roughly 200 K. Low-temperature measurements of both the specific heat and the spin susceptibility show no anomalies that suggest a phase transition or a spin-freezing temperature.

One may ask whether there are more direct probes of the QSL. In fact, we have not yet discussed one of the defining properties of QSLs: They possess a high degree of quantum entanglement. Recall the picture of the QSL state as a superposition of all possible arrangements of singlets that cover the lattice, as shown in figures 1d and 1g. As a highly entangled state, the QSL cannot be written as a product state of finite spatial blocks of singlets. Consequently, the fundamental excitations may be described by fractionalized quantum numbers. For a more detailed discussion on spin excitations, see the box on page 33.

The fractional quantum Hall state, in which the charge degrees of freedom have fractional statistics, is the one known experimental realization of such a highly entangled state. The QSL is a leading candidate to become the second known experimental realization.

The specific heat in the organic triangular-lattice compounds shows a large term that is linear in T below 1 K, common for a metal with a Fermi surface but very unusual for a magnetic insulator such as EtMe$_3$Sb[Pd(dmit)$_2$]$_2$. In addition, the spin susceptibility appears to saturate in the $T \to 0$ limit at a value consistent with having spin excitations that behave as fermions.

Measuring thermal conductivity is another way to investigate spin excitations. In a magnetic insulator, the thermal conductivity contains contributions from phonons, magnetic excitations (spinons or magnons), and other propagating excitations. At low temperatures, the thermal conductivity of EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ has a large term linear in T,[10] as shown in figure 2b. As with specific heat, the linear term indicates free fermions are the carriers of heat, which is surprising for an insulator.

The T-linear term is as large as the contribution by conduction electrons in metallic brass, so the mean free path for the heat-carrying quasiparticles must be exceedingly long—on the order of 1 μm. Such a large T-linear component in the thermal conductivity lends further credence to the idea that the spin excitations behave like fermions.

The persuasive power of those measurements on the putative organic QSLs lies in the fact that they go beyond simply showing the absence of ordering and freezing. They show that the materials' properties have specific temperature dependences that are consistent with spin excitations behaving as nearly free fermions rather than as magnons.

Another difficulty that arises in definitively proving the existence of a QSL in a particular material is that a vast array of theoretical QSL states are possible, each with slightly different properties.[6] Some QSLs have a so-called spin gap between the ground state and the first excited state. Others, such as the proposed state for the organics, are gapless.

Hence one of the best ways to make progress is to find a material with a theoretically tractable spin Hamiltonian and study the spin excitations directly with spin-sensitive probes. In the past several years, exciting developments have been made on both fronts for the Heisenberg model on the kagome lattice.

Advances in numerical calculations have shed much light on the possibility that the ground state of the $S = ½$ kagome antiferromagnet is a QSL. Density-matrix renormalization-group studies suggest that there is a spin gap.[11] Further support comes from calculations showing nonzero topological entanglement entropy for the model. However, the difficulty in performing calculations for the quantum kagome antiferromagnet is highlighted by other theoretical studies, such as variational wave-function calculations,[12] that show different gapless spin liquids can be very competitive as ground states.[6]

Herbertsmithite

Concomitant with the recent theoretical progress, experimental studies have identified one particularly promising $S = ½$ kagome lattice material: the mineral herbertsmithite. Given by the chemical formula ZnCu$_3$(OH)$_6$Cl$_2$, herbertsmithite has a crystal structure, shown in figure 3a, that is composed of sheets of Cu^{2+} ions in an ideal kagome geometry with a layer

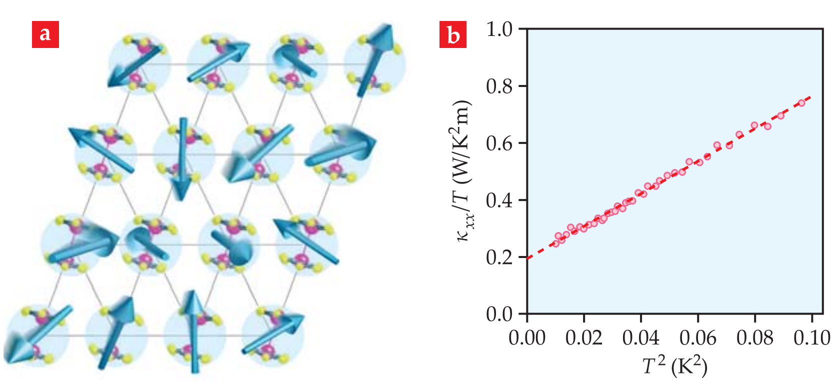


FIGURE 2. ORGANIC QUANTUM SPIN LIQUID CANDIDATES. (a) This depiction of the two-dimensional triangular planes in the organic compound EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ illustrates how pairs of Pd(dmit)$_2$ molecules (highlighted in the blue circles) each share an unpaired, spin-½ electron denoted by the blue arrow. **(b)** As a function of the square of the temperature T^2, the thermal conductivity κ_{xx} divided by T extrapolates to a finite intercept at $T = 0$ K. That indicates a large T-linear contribution, consistent with spinons with a Fermi surface. (Adapted from ref. 10.)

of nonmagnetic Zn^{2+} and Cl^- ions in between. Early studies on powder samples confirmed the lack of magnetic order or spin freezing down to temperatures below 0.001 J/k_B ($J/k_B \approx 200$ K for herbertsmithite).[13]

Importantly, single-crystal samples can be grown, and they have been used for various detailed measurements. We will highlight two recent experiments based on neutron scattering and NMR that shed valuable light on the nature of the possible QSL ground state.

As discussed in the box, a signature feature of QSLs is that they support exotic spin excitations called spinons that carry fractional quantum numbers. To obtain detailed information about the fractionalized spinon excitations requires an energy- and momentum-resolved technique. Single-crystal inelastic neutron scattering is a powerful probe of that information. The magnetic neutron-scattering cross section is directly proportional to the dynamic structure factor $S(\mathbf{Q}, \omega)$, where $\mathbf{Q}$ and ω are the momentum and energy transferred to the sample, respectively. Thus the scattered neutron intensity is a measure of $S(\mathbf{Q}, \omega)$, the Fourier transform, in time and space, of the spin–spin correlation function.

Plots of $S(\mathbf{Q}, \omega)$ for herbertsmithite are shown[14] in figure 3b. Surprisingly, the scattered intensity is exceedingly diffuse, spanning a large fraction of the hexagonal Brillouin zone, the unit cell in reciprocal space, even at a temperature that is two orders of magnitude below J/k_B. That indicates the lack of any tendency for ordering and is in strong contrast to observations in nonfrustrated quantum magnets. For example, the square-lattice antiferromagnet La_2CuO_4 develops substantial antiferromagnetic correlations for $T < 0.5\ J/k_B$, even in the paramagnetic state, and low-energy neutron scattering becomes strongly peaked in reciprocal space.[5] In herbertsmithite, the scattered intensity is not strongly peaked at any specific point. That behavior is also markedly different from what is observed in the larger-spin $S = 5/2$ kagome antiferromagnet $KFe_3(OH)_6(SO_4)_2$, which orders magnetically at low temperatures. There, quasielastic scattering peaks develop above the ordering temperature.[15]

The neutron data in figure 3b also show that the scattered signal is broad in energy. Hence neutron scattering provides direct evidence that at low temperatures, the spin excitations in herbertsmithite form a continuum, in contrast to the conventional spin waves expected in ordered antiferromagnets (see the box). Such a continuum, a signature of fractional spin excitations, has so far been clearly observed only in 1D systems. The measurement serves as a hallmark of the QSL state in herbertsmithite. By integrating the inelastic scattering over energy, one can extract information on the instantaneous spin correlations. The pattern of intensity in reciprocal space (in particular, the green rings of scattering at 2 meV and 6 meV in figure 3b) indicates that the spins are dominated by short-range singlet correlations, consistent with Anderson's RVB conjecture.

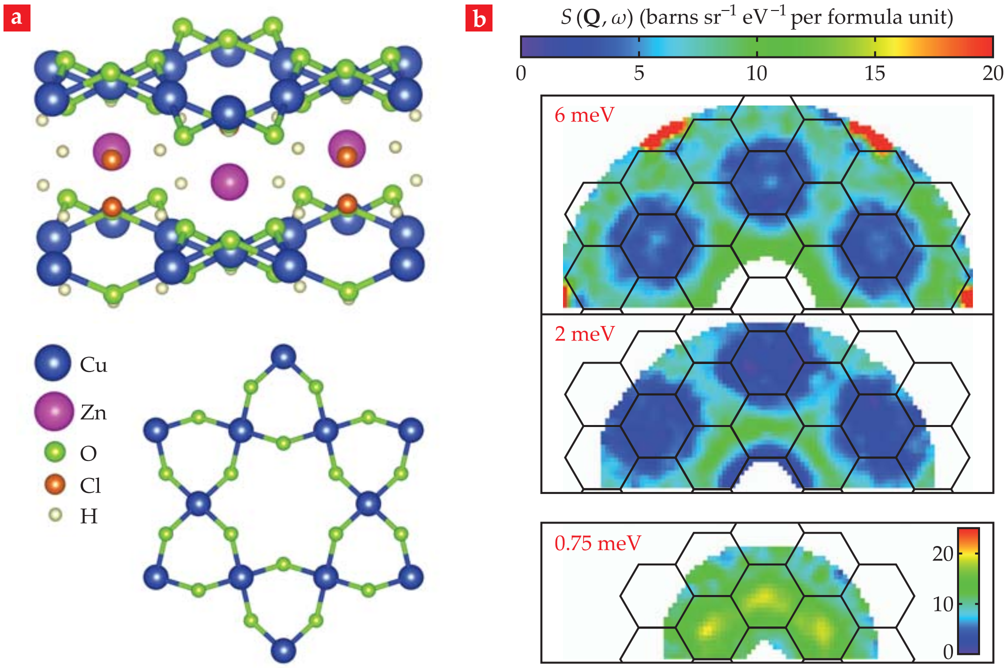
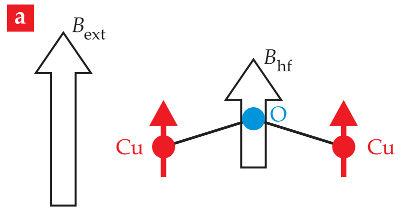


FIGURE 3. THE KAGOME LATTICE as host for quantum spin liquid. **(a)** The crystal structure of the mineral herbertsmithite consists of copper and oxygen ions in kagome planes that are separated by nonmagnetic zinc, chlorine, and hydrogen interlayer ions. The bottom picture shows the two-dimensional structure of the kagome plane. **(b)** Inelastic neutron scattering measurements, made at 1.6 K on a single-crystal sample, map the spin excitations. The reciprocal-lattice unit-cell boundaries, drawn in black, provide orientation in reciprocal space. The signal at 0.75 meV is dominated by impurity scattering. However, plots of the dynamic structure factor $S(\mathbf{Q}, \omega)$ for energies of 2 meV and 6 meV reveal broad rings of scattering (green). The pattern is consistent with the ground state spin arrangement shown in figure 1g. In addition, the scattering pattern's insensitivity to energy indicates a continuum of spin excitations. Both behaviors suggest herbertsmithite is a quantum spin liquid. (Adapted from ref. 14.)

To further specify the ground state of the kagome Heisenberg antiferromagnet, one must probe the low-energy sector of spin excitations. In herbertsmithite, however, the nonmagnetic Zn^{2+} sites located between the kagome planes are occupied, with about 15% probability, by extra Cu^{2+} ions.[16] Those impurities also have spin $S = 1/2$ but are not strongly bound to the intrinsic Cu^{2+} kagome network, and the energy scale of the interaction between defect spins is small. Accordingly, the magnetic response of the defects is concentrated at low energies. Thus the impurity response begins to dominate the neutron scattering intensity for energy transfers below 2 meV, as seen by the different pattern of the intensity at 0.75 meV in figure 3b, and masks the intrinsic behavior of the kagome lattice. That also means the impurity contribution dominates the bulk-averaged spin susceptibility, deduced from magnetization measurements, at low temperatures,[13] which makes it difficult to test for the presence of a spin gap.

Because NMR is a local probe, it can separately detect nuclear spins that are far from defects or near defects. That ability makes it ideally suited for investigating the intrinsic low-energy



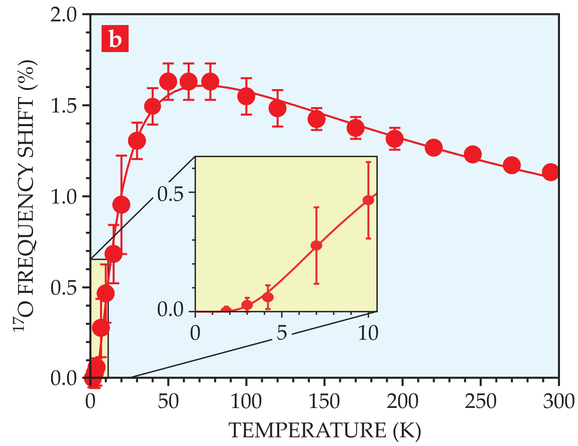


FIGURE 4. INTRINSIC SPIN SUSCEPTIBILITY of herbertsmithite. **(a)** An external magnetic field B_{ext} induces polarization of the Cu^{2+} sites (red). Covalent effects transfer that polarization to the O^{2-} site (blue) and exert a hyperfine magnetic field B_{hf} on the ^{17}O nuclear spin that is proportional to the intrinsic spin susceptibility χ_{kagome}. **(b)** NMR measures the resonant absorption of RF waves between nuclear spin levels split by the total magnetic field $B_{ext} + B_{hf}$. The behavior of χ_{kagome} in herbertsmithite is measured as the ^{17}O NMR frequency shift in $B_{ext} = 3.2$ T. The inset shows that χ_{kagome} asymtotically goes to zero below temperature $T \approx 0.03\ J/k_B$ (J is the magnetic exchange energy and k_B is Boltzmann's constant), signaling the formation of a collective singlet ground state. (Adapted from ref. 17.)

spin excitations in the presence of defects. Recent NMR studies of herbertsmithite single crystals enriched in oxygen-17 or deuterium enabled measurements of intrinsic and defect-induced local spin susceptibilities (χ_{kagome} and χ_{defect}, respectively)[17] down to $T \approx 0.01\ J/k_B$.

In condensed-matter NMR measurements, one detects the resonant absorption of RF waves at the Zeeman frequency $\omega_0 \approx \gamma_n B_{ext}$ of the nuclear spin in an external magnetic field B_{ext} (γ_n is the nuclear gyromagnetic ratio of the observed nuclear spins). But in herbertsmithite, B_{ext} also polarizes the Cu^{2+} magnetic moments in proportion to χ_{kagome}. The polarized moments, in turn, exert an additional effective hyperfine magnetic field B_{hf} on adjacent nuclear spins, as illustrated in figure 4a; B_{hf} shifts the resonant frequency by $\gamma_n B_{hf}$. Because B_{hf} at ^{17}O sites that are far from defects is proportional to χ_{kagome}, one can determine the behavior of χ_{kagome} through measurements of the relative NMR frequency shift, $B_{hf}/B_{ext} \propto \chi_{kagome}$.

In contrast, B_{hf} induced by defects at nearby ^{17}O and ^2D sites is proportional to χ_{defect}, which has a $1/T$ temperature dependence and is strongly negative at the crucial low-temperature region. As a consequence, ^{17}O and ^2D NMR signals arising from the immediate vicinity of the defects are separated from the main NMR signals arising from the defect-free parts of the crystal. Because the split-off defect-induced NMR peak does not mask the intrinsic ^{17}O NMR peak, the presence of impurity spins at the Zn^{2+} sites does not hamper NMR measurements of χ_{kagome}.

The temperature dependence of the frequency shift,[17] shown in figure 4b, reveals that χ_{kagome} decreases at low temperatures, and asymptotically approaches zero below 6 K (approximately 0.03 J/k_B). The vanishing of χ_{kagome} implies that Cu^{2+} electron spins form a collective spin-singlet state, smoking-gun evidence for a QSL ground state such as the one illustrated in figure 1g and a spin gap $\Delta \approx 0.03\ J$ between the ground and lowest excited states. That value is comparable to numerical prediction based on the density-matrix renormalization-group techniques.[11]

Tip of the iceberg

The few example materials that we have discussed are hopefully just the first among many to be discovered that possess a QSL ground state. Beyond materials based on triangular and kagome lattices, other exciting new avenues for finding spin liquids involve highly frustrated pyrochlore lattices composed of corner-sharing tetrahedra with highly anisotropic interactions, and honeycomb lattices with bond-dependent interactions.

In the decade since PHYSICS TODAY's report on QSLs (February 2007, page 16), significant progress has been made on both theoretical and experimental fronts. Specific characteristics of the excitation spectrum can now be predicted and measured in model systems. Such fruitful interplay between theory and experiment allows one to seriously address the titular question of this article, "Do quantum spin liquids exist?" A search that began with Anderson's notion more than four decades ago is now approaching a final conclusion. Of course, a high bar of evidence is rightly required for a definitive answer. The community has yet to achieve consensus on the issue, but in our opinion, the continuing stream of evidence points toward an answer in the positive.

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