Spin jam induced by quantum fluctuations in a frustrated magnet

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Since the discovery of spin glasses in dilute magnetic systems, their study has been largely focused on understanding randomness and defects as the driving mechanism. The same paradigm has also been applied to explain glassy states found in dense frustrated systems. Recently, however, it has been theoretically suggested that different mechanisms, such as quantum fluctuations and topological features, may induce glassy states in defect-free spin systems, far from the conventional dilute limit. Here we report experimental evidence for existence of a glassy state, which we call a spin jam, in the vicinity of the clean limit of a frustrated magnet, which is insensitive to a low concentration of defects. We have studied the effect of impurities on SrCr9Ga12−pGa+pO19 (SCGO(p)), a highly frustrated magnet, in which the magnetic Cr3+ (s = 3/2) ions form a quasi-2D triangular system of bipyramids. Our experimental data show that as the nonmagnetic Ga3+ impurity concentration is changed, there are two distinct phases of glassiness: an exotic glassy state, which we call a spin jam, for the high magnetic concentration region (p > 0.8) and a cluster spin glass for lower magnetic concentration (p < 0.8). This observation indicates that a spin jam is a unique vantage point from which the class of glassy states of dense frustrated magnets can be understood.

Significance

We report experimental evidence for a glassy state induced by quantum fluctuations, a spin jam, that is realized in SrCr9Ga12−pGa+pO19 [SCGO(p)], a highly frustrated magnet, in which the magnetic Cr3+ (s = 3/2) ions form a quasi-two-dimensional triangular system of bipyramids. Our new experimental data and our theoretical spin jam model provide, for the first time, to our knowledge, a coherent understanding of the existing experimental data of this fascinating system. Furthermore, our findings strongly support the possible existence of purely topological glassy states.


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The interpretation of the frozen state below Tf is still controversial. One possibility suggested was a spin liquid with unconfined spinons or resonating valence bond state, based on NMR and µSR studies (15, 16). Many-body singlet excitations were also suggested to be responsible for the Cχ ∝ T2 behavior (14).

Recently, some of us presented an alternative scenario involving a spin jam state by considering the effects of quantum fluctuations in the disorder-free quasi-2D ideal SCGO lattice with a simple nearest neighbor (NN) spin interaction Hamiltonian $H = J \sum_{i,j} S_i \cdot S_j$ (18, 19). The spin jam framework provided a qualitatively coherent understanding of all of the low-temperature behaviors such as that a complex energy landscape is responsible for the frozen state without long-range order (18), and Halperin–Saslow (HS)-like modes for the Cχ ∝ T2 and χ(1)/ω behaviors (5, 18). In this system, which we refer to as the ideal SCGO model (iSCGO), semiclassical magnetic moments (or spins) are arranged in a triangular network of bipyramids and interact uniformly with their NN (18, 19). The microscopic mechanism for the spin jam state is purely quantum mechanical. The system has a continuous and flat manifold of ground states at the mean field level, including locally collinear, coplanar, and non-coplanar spin arrangements. Quantum fluctuations lift the classical ground state degeneracy (order by fluctuations), resulting in a complex rugged energy landscape that has a plethora of local minima consisting of the locally collinear states separated from each other by potential barriers (18). Although the work of ref. 18 dealt with a similar phase space constriction by quantum fluctuations as the aforementioned other theoretical works did, we would like to stress here the difference between the two: Whereas the other works mainly focused on the selection of the long-range-ordered (LRO) energetic ground state, the work of ref. 18 showed a spin jam state at low temperatures.
that the short-range-ordered (SRO) states that exist at higher energies are long-lived, dominate entropically over the LRO states, and govern the low-T physics.

The introduction of nonmagnetic impurities into a topological spin jam state breaks some of the constraints in the system, and possibly allows local transitions between minima, with a time scale dependent on the density of impurities. At a sufficiently high vacancy concentration, the system exits the spin jam state and becomes either paramagnetic or an ordinary spin glass at lower temperatures. Here we try to identify and explore the spin jam regime in an experimentally accessible system. The three most important signatures we seek for the existence of a spin jam state, different from conventional spin glass states, are (i) a linear dependence of the imaginary part of the dynamic susceptibility at low energies, \( \chi''(\omega) \propto \omega \), (ii) intrinsic short range static spin correlations, and (iii) insensitivity of its physics to nonmagnetic doping near the clean limit. In the rest of the paper, we provide experimental demonstration of these properties.

Experimentally, there are, so far, two materials, SrCr\(_9\)Ga\(_{12}\)O\(_{19}\) [SCGO(j)] (1–3, 13–17, 20) and q-ferrites like Ba\(_3\)Sn\(_4\)ZnGa\(_{3}\)Cr\(_{2}\)O\(_{22}\) (BSZGCO) (21), in which the magnetic Cr\(^{3+} \) (3d\(^3\)) ion surrounded by six oxygen octahedrally, form distorted quasi-2D triangular lattice of bipyramids (20, 21) as shown in Fig. 1A, and thus may realize a spin jam state. We would like to emphasize that these systems are very good insulators (resistivity \( \rho > 10^{13} \) Ω-cm at 300 K) and the Cr\(^{3+} \) (t\(_{2g}\)) ion has no orbital degree of freedom. Furthermore, the neighboring Cr ions share one edge of oxygen octahedral, and thus the direct overlap of the t\(_{2g}\) orbitals of the neighboring Cr\(^{3+} \) ions make the AFM NN Heisenberg exchange interactions dominant and further neighbor interactions negligible (22, 23), as found in Cr\(_2\)O\(_3\) (24) and ZnCr\(_2\)O\(_4\) (25).

**Experimental Data**

We have performed elastic and inelastic neutron scattering that directly probe spin–spin correlations and bulk susceptibility measurements on SCGO(p) with various values of \( p \) over \( 0.2 \leq p \leq 1.0 \) spanning almost the entire region of \( p \). To first characterize the samples and to construct the \( T-p \) phase diagram, we have performed dc magnetic susceptibility, \( \chi_{\text{bulk}} \), and elastic neutron measurements. The data obtained from two samples with \( p = 0.968(6) \) and 0.620(8), both of which are well above the percolation threshold for the triangular lattice of bipyramids, \( p_\text{c} \approx 0.5 \) (9), are shown in Fig. 2A and B, respectively (the data obtained from samples with other \( p \) values are shown in Fig. S3). For both samples, \( \chi_{\text{bulk}} \) exhibits similar FC and ZFC hystereses below \( T_\text{f} = 3.68 \text{K} \) \( \{ p = 0.968(6) \} \) and 1.76 K \( \{ p = 0.620(8) \} \) that are much lower than their large Curie–Weiss temperatures \( \Theta_{\text{CW}} \approx 504 \text{K} \) \( \{ p = 0.968(6) \} \) and 272 K \( \{ p = 0.620(8) \} \) (see Fig. S1). The high frustration index \( f = (\Theta_{\text{CW}}/T_\text{f}) \approx 130 \) indicates the presence of strong frustration in both systems. Elastic neutron scattering intensity with an instrumental energy resolution of \( \hbar\omega \leq 25 \mu \text{eV} \) starts developing at temperatures higher than their \( T_\text{f} \) determined by \( \chi_{\text{bulk}} \), as is expected for spin freezing for measurements with different energy resolution (26). Fig. 1B summarizes the results obtained from all of the samples studied.

We observe strikingly different behaviors in the low-energy spin dynamics of the \( p = 0.968(6) \) and 0.620(8) despite the similar temperature dependences of \( \chi_{\text{bulk}} \) and of elastic neutron scattering intensity. As shown in Fig. 2C, in the frozen state of \( p = 0.968(6) \), the inelastic neutron scattering intensity \( I(Q,\hbar\omega) \) is weak at very low energies below 0.25 meV and gets stronger as \( \hbar\omega \) increases. In contrast, in the frozen state of \( p = 0.620(8) \), \( I(Q,\hbar\omega) \) is stronger at very low energies below 0.25 meV and gets weaker as \( \hbar\omega \) increases (see Fig. 2D). This stark difference hints that the frozen states of \( p = 0.968(6) \) and 0.620(8) are different in nature.

To investigate more carefully the difference between the low and high doping regions, we have also performed time-of-flight neutron scattering measurements on several samples with various doping concentration from \( p = 0.968(6) \) to 0.459(5) (see Fig. S4). The data exhibit a continuum spectrum centered in the vicinity of \( Q_{\text{max}} \approx 1.5 \text{ Å}^{-1} \) that corresponds to \((2/3,2/3,1.8)\) confirming that the kagome–triangular–kagome trilayer is responsible for the low-energy dynamic spin correlations (19). This resembles the

**Fig. 1.** (A) In SrCr\(_9\)Ga\(_{12}\)O\(_{19}\) [SCGO(j)], the magnetic Cr\(^{3+} \) (3d\(^3\), \( s = 3/2 \)) ions form the kagome–triangular–kagome trilayer (Top). The blue and red spheres represent kagome and triangular sites, respectively. When viewed from the top of the layers, they form the triangular network of bipyramids (Bottom). (B) The \( p-T \) phase diagram of SCGO(p) constructed by bulk susceptibility and elastic neutron scattering measurements on powder samples with various \( p \) values. The freezing temperatures, \( T_\text{f} \), marked with blue square and black circle symbols are obtained by bulk susceptibility and elastic neutron scattering measurements, respectively. Note that the values of \( T_\text{f} \) are much lower than the Curie–Weiss temperatures (see Fig. S1). Filled blue squares represent the data obtained from samples whose crystal structural parameters including the Cr/Ga concentrations were refined by neutron diffraction measurements (see Fig. S2 and Tables S1–S4), and open blue squares represent samples with nominal \( p \) values. For nominal \( p = 0.2 \), no freezing was observed down to 50 mK (see Fig. S3).

**Fig. 2.** (A and B) \( T \) dependences of bulk susceptibility (blue solid squares) and elastic neutron scattering intensity (black open circles) for SCGO(p) with (A) \( p = 0.968(6) \) and (B) \( p = 0.620(8) \). Bulk susceptibility was measured with an application of external magnetic field \( H = 0.01 \text{ Tesla} \). The black dashed lines indicate the nonmagnetic background for the elastic neutron scattering. (C and D) Contour maps of neutron scattering intensity as a function of momentum and energy transfer for (C) \( p = 0.968(6) \) and (D) \( p = 0.620(8) \) measured at \( T = 1.4 \text{ K} \). The neutron scattering measurements were performed with incident neutron wavelength of 6 Å. Intensities were normalized to an absolute unit by comparing them to the \((0,0,2)\) nuclear Bragg peak intensity.
energy continuum expected for spin liquids or cooperative paramagnets; however, in contrast to a spin liquid, in our system, static spin correlations develop below $T_f$ as well. The $Q$ dependence of the elastic magnetic neutron scattering intensity, $I_{neut}^{el}(Q) = I_{el}(Q, 1.4 K) - I_{el}(Q, 20 K)$, was obtained with an energy window of $|\hbar Q| \leq 25$ μeV, where the subtraction of the signal above the magnetic transition eliminates background from sources not related to the transition. As shown in Fig. 3, for $p = 0.968(6)$, very close to the clean limit, $I_{neut}^{el}(Q)$ exhibits a broad peak at $Q_{max} = 1.49$ Å$^{-1}$. Its broadness indicates the short-range nature of the static spin correlations. Surprisingly, upon increasing nonmagnetic impurity concentration up to about 20% ($p \sim 0.8$), the shape of $I_{neut}^{el}(Q)$ remains the same. Only upon further doping does $I_{neut}^{el}(Q)$ become broader, and the peak position $Q_{max}(p)$ starts shifting down.

To quantitatively investigate the correlation length $\xi(p)$ and peak position $Q_{max}(p)$ as a function of $p$, we fit $I_{neut}^{el}(Q)$ to a simple Lorentzian function for each $p$, $I_{neut}^{el}(Q) \propto 1/[\text{HWHM}^2 + (Q - Q_{max})^2]$ where HWHM is the half width at half maximum of $I_{neut}^{el}(Q)$. As a measure of the static spin correlation length scale of the frozen state, we can use $\xi_{SWTHM} = 1/\text{HWHM}$. Note that the powder averaging would introduce extrinsic broadening to $I_{neut}^{el}(Q)$, and thus $\xi_{SWTHM}$ is underestimated, in comparison with the correlation length determined from single-crystal data, $\xi_{SWTHM} = 4.6(2)$ Å for SGCO ($p = 0.067$) (19). However, the $p$ dependence of $\xi_{SWTHM}$ serves our search for an intrinsic state.

The resulting $\xi_{SWTHM}$ and $Q_{max}$ are shown in Fig. 3B. Remarkably, both $\xi_{SWTHM}$ and $Q_{max}$ exhibit a flat behavior near the clean limit up to 20% doping, which is a direct evidence for existence of a distinct phase over $1 < p < 0.8$ where the intrinsic short-range static correlations are independent of nonmagnetic doping. This can be naturally described as a spin jam state at the clean limit. A spin jam state can be intrinsically short range, and has an intrinsic static correlation length $\xi_{int}$. Thus, when nonmagnetic doping is low and the typical distance between the nonmagnetic impurities is larger than $\xi_{int}$ the spin correlations are not affected. Only upon significant doping would the spin correlations get disturbed to make $\xi(p)$ shorter than $\xi_{int}$ when typical distance between impurities is short.

If the $p > 0.8$ phase is distinct from the lower $p < 0.8$ phase, then the low-energy spin excitations in those two phases should have different characteristics. To see this, we show in Fig. 4 the imaginary part of the dynamic susceptibility $\chi''(\omega) = \frac{\pi}{\hbar^2} \Gamma(\omega,\hbar\omega)[1 - e^{-\hbar\omega/k_B T}]$ and $k_B$ is the Boltzmann factor. For $p = 0.968(6)$, upon cooling in the cooperative paramagnetic state from 20 K (< $Q_{max}$) to 5 K ($\sim T_f$), $\chi''(\omega)$ changes from being linear to almost flat (see Fig. 4A). The nearly $\omega$-independent low-energy $\chi''$ implies that a distribution of the characteristic spin relaxation rates, $\Gamma$, is present, which is common in glassy transitions (27). A distribution of $\Gamma$ would yield $\chi''(\omega) \propto \frac{\Gamma_{min}}{\hbar\omega^{2}} + \frac{\Gamma_{max}}{(\alpha/\omega + \Gamma_{max})^{2}}$ where a term, $\tan^{-1}(\omega/\Gamma_{min})$, is ignored since $\Gamma_{max} \gg 1$ meV, much larger than the $\omega$-range of interest. Fitting the $T = 6$ K ~ $7$ K data to the model yields a distribution of $\Gamma$ with $\Gamma_{min} = 0.053(4) \text{meV}$ (see the red solid line in Fig. 4A).

Upon further cooling into the frozen state, however, $\chi''(\omega)$ exhibits hardening: The weight gets depleted and becomes linear at low energies (see the 1.4 K data), which is consistent with a previous neutron scattering study of SGCO [$p = 0.92(5)$] (17). The 1.4 K data can still be fitted to $\chi''(\omega) \propto \frac{\Gamma_{min} \omega^{2}}{\omega^{2} + \Gamma_{max}}$, with $\Gamma_{max} = 0.25(3) \text{meV}$, which indicates that a distribution of $\Gamma$ is still present but with the larger minimum cutoff than that of 6 K. Also, for $\omega \ll 2\Gamma_{min}$, $\chi''(\omega)$ is linear with $\omega$.

For $p = 0.459(5)$, on the other hand, we observe a fundamentally different behavior in the frozen state: As shown in Fig. 4F, below freezing, rather than hardening, the spectral weight at low frequencies $\chi''(\omega)$ exhibits a prominent increase peaked at ~0.2 meV. Obviously, the data cannot be reproduced by $\tan^{-1}(\omega/\Gamma_{min})$ alone. Instead it behaves more like a Lorentzian with one characteristic relaxation rate, $\alpha/\omega + \Gamma_{max}$.

The clear difference in the behavior of $\chi''(\omega)$ in the frozen state between the two regimes of $p$ is another evidence that the frozen state in the vicinity of the clean limit is indeed a distinct state. To see how the evolution of the state occurs as a function of $p$, we have fitted $\chi''(\omega)$ measured in the frozen state of each $p$ to the sum of the two contributions, $\chi''(\omega) \propto \frac{\Gamma_{min} \omega^{2}}{\omega^{2} + \Gamma_{max}} + f \cdot \frac{\Gamma_{min} \omega^{2}}{\omega^{2} + \Gamma_{max}}$, where $f = 0.05(6)$ for $p = 0.968$ and $f = 0.06(5)$ for $p = 0.917$. As $p$ decreases further, i.e., as nonmagnetic doping increases, however, $f$ gradually increases. As shown in Fig. 4C and D, the $\tan^{-1}mittel$ (dotted line) dominates for $p \geq 0.777(6)$, whereas $\tan^{-1}mittel$ is very small for $p = 0.968$.

Fig. 3. (A) $Q$ dependence of the elastic magnetic scattering intensity measured for various values of $p$ at 1.4 K, except for $p = 0.459(5)$ at $T = 0.27$ K. Nonmagnetic background was determined at 20 K and subtracted. Solid lines are fits to a simple Lorentzian. Dashed lines and arrows represent the fitted FWHM and peak positions, respectively. (B) The peak position and the power static spin correlation length that were obtained from the fits are plotted as a function of $p$. (C) The fraction of the contribution from the single Lorentzian to the dynamic susceptibility, as shown in Fig. 4, is plotted as a function of $p$.
the Lorentzian term (dashed line) dominates for $p \lesssim 0.7$. The $f(p)$ over a wide range of $p$ is plotted in Fig. 3C. This confirms, upon doping, a crossover from a frozen state near the clean limit to another frozen state at high nonmagnetic concentration limit.

What is then the nature of the frozen state in the vicinity of the clean limit? The hardening and linear behavior of the low-energy spin fluctuations in the frozen state of SCGO ($p > 0.8$) can be explained as HS-type modes in a spin jam (5, 28, 29). HS modes are long-range collective modes that may be viewed as an analog of the Goldstone modes associated with continuous symmetry breaking in systems without long-range order (5). In contrast, for low values of $p$, as defect concentration is increased, the low-energy spectral weight is eventually dominated by contributions from a distribution of local spin clusters. This is consistent with the previous specific heat data that reported the $C_s \propto T^2$ behavior robust against dilution for $p \gtrsim 0.8$ (14), whereas, for $p \lesssim 0.8$, the exponent starts decreasing with decreasing $p$ (30).

As further quantitative evidence of an HS mechanism for the specific heat, $C_s \propto T^2$ may be roughly estimated for a spin jam. HS theory with a dispersion $\epsilon_{r a n d} \approx \sqrt{\rho_i / \bar{\rho}}$ gives a contribution to specific heat $C_v (k_B T)/T^{1/2} \approx [\pi (3/2) K_B T]/(\pi h^2 \gamma_r^2)$, where $\gamma_r$ is the gyromagnetic ratio, $\langle 3 \rangle \approx 1.2$ is the Riemann Zeta function, and we have accounted for three modes for isotropic spins. Using experimental values of $\bar{\rho} = 0.007$ electromagnetic units (emu) per mole Cr (see Fig. 2A) and using the HS upper bound estimate $\bar{\rho} = (J/H V) \sum_{r a n d} (\langle S_i \rangle \cdot \langle S_j \rangle)$ adapted to iSCGO, $J = 9$ meV, and $\rho_i = \frac{\left[2J^2 \cdot \left(18 \sum_{a b c d} S_2 \cdot S_2\right)\right]}{\gamma_r (3)}$, taking $\langle S_i \rangle = 0.95\mu_B$/Cr from ref. 17, we get a theoretical estimate of $(C_v/T^2) \approx 0.07$ Joulles per mole Cr. $K^{-1}$, which is consistent with the experimental value of $\sim 0.059$ Joulles per mole Cr. $K^{-1}$.

**Discussion.**

The salient features observed in SCGO ($p > 0.8$) are consistent with the recent understanding of the iSCGO case with $J' = J$ spin jam (18), where $J$ and $J'$ are the intralayer and interlayer Heisenberg NN couplings, respectively. For real SCGO samples, $J' < J$, in general, due to lattice distortion. Furthermore, there are two different $J$ in the kagome plane: $J_1$ and $J_2$ for the bonds within each bipiramidal and for the bonds of the linking triangle connecting the bipyramids. However, at the mean field level, the case of $J_1 = J_2 = J$ yields the same ground states as the uniform case of $J_1 = J_2 = J$ because the ground state of the uniform case satisfies both the AFM constraints of the linking triangle and the bipyramid. Thus, we limit our discussion to the case of $J_1 \neq J_2 \neq J$. For these situations, classical magnetic ground states have been described as a function of $J/J'$ in ref. 19. These can be obtained from the $J = J'$ case by coherent rotations of a subset of the spins. The classical manifold of ground states remains degenerate for $J' < J$, as can be seen from the flat zero-energy bands that are present in the linear spin wave analysis around long-range ordered states described in Fig. S5. As the ratio $J'/J$ changes from 1 to 0, the system moves from the ideal trilayer to decoupled kagome layers (accompanied by a layer of noninteracting spins), at which point local zero-energy modes, such as weathervane modes (7), associated with kagome physics, appear. For the isolated semiclassical kagome, an extensive configurational entropy of low minima appears with kinetic barriers associated with the weathervane motion. At low enough temperatures, tunneling is suppressed and the system freezes. On general grounds, local modes such as the weathervane will become delocalized when the layers are coupled, suggesting that for $J \neq J'$, both the barriers between local minima remain nonlocal in nature and thus freezing is more effective than for the decoupled kagome.

The quantum fluctuations-induced spin jam scenario is consistent with the system freezing at temperatures much lower than $\Theta_{CW}$. An energy scale for spin fluctuations in the clean limit can be determined from the potential barrier between local (ordered) minima and is given by $E_{SCGO} \approx 0.05J$ (18). For SCGO ($p > 0.8$), $E_{SCGO}/k_B \approx 7.8$ K, which is close to the experimentally determined $T_r \approx 4.4 \pm \Theta_{CW} \approx 500$ K.

One may consider anisotropic interactions such as the Dzyaloshinskii–Moriya (DM) as the origin for the spin freezing. We believe that a simple DM interaction will not generate such a complex energy landscape and cannot explain coherently all of the experimental data as the quantum fluctuation-induced spin jam scenario does. However, theoretical confirmation about such an anisotropic scenario is needed, which is beyond the scope of this paper.

Upon weak nonmagnetic doping, the complex energy landscape is modified and the kinetic barriers become finite; however, the overall picture remains the same. Further doping will weaken the order by fluctuations mechanism and the selection of coplanar states, and a different glassy state will emerge. Indeed, our observation of a crossover as a function of doping is consistent with Henley (9), who, remarkably, speculated on the possibility of a defect-induced crossover as a function of doping, close to the clean limit, from a nongeneric phase (dominated by coplanar states) to a generic phase in large-spin kagome and SCGO systems. It is worthwhile to discuss here the concept of jamming in a broader context. In granular systems, a transition occurs into a special type of glassy state called jammed state with the increase of the density of constituents due to a complex energy landscape (9). In analogy, one may view the approach of the disorder-free frustrated magnets as the addition of system constituents, i.e., removal of nonmagnetic impurities, and the state of the clean system as the “spin-jammed” state. In such a system, a large set of mostly irregular spin configurations form local minima, the transition between which requires the simultaneous reorientation of a large number of spins. The recent theoretical work of ref. 18 has shown how a disorder-free quasi-2D frustrated magnet with a simple NN spin interaction Hamiltonian is a frozen spin jam of topological nature, at low temperatures. It was also established that the locally collinear states differ by collective reorientations of spins, where the smallest units of the mean field zero-energy excitations are extended along lines, called spaghetti modes. The extended nature of the reorientations is responsible for particularly large dynamical barriers, of topological nature, to tunneling between minima (18). The effect of an order by fluctuations may also be a possible mechanism for the glassy state of the systems (6, 7, 13). With less robust nature than in the iSCGO (9), as the presence of local tunneling through weathervane modes involving six spins may hasten approach to a global ground state.

In comparison, surprisingly, the 3D pyrochlore $Y_2Mo_2O_7$ also exhibits a similar $C_s \propto T^2$ behavior at low temperatures but with $D$ less than the value of 3 expected for a 3D system (34). The magnetic Mo$^{4+}$ (4d$^0$) ions form a 3D network of corner-sharing tetrahedra. If these magnetic moments are isotropic, and antiferromagnetically and uniformly interacting with their NN only, the system is supposed to yield the highest degree of frustration. One may speculate that the freezing in $Y_2Mo_2O_7$ may also be explained in terms of a spin jam; however, we would like to point out crucial differences between the systems. In particular, the frustration index of $Y_2Mo_2O_7$ is two orders of magnitude smaller than in SCGO: $f \approx 0.087/0.45/20K/2.3$. This can be understood by the facts that, unlike SCGO, which is an excellent insulator, $Y_2Mo_2O_7$ is semiconducting ($\rho \sim 10^{-2}$ $\Omega \cdot cm$ at 300 K), and the neighboring Mo ions share one corner of oxygen octahedra, which tends to result in nonnegligible longer-range magnetic interactions (22). Moreover importantly, the magnetic Mo$^{4+}$ ($t_2^g$) ions are orbitally degenerate (34, 35); it is well known that orbital degeneracy has a great tendency to modify the nature of a magnetic network, and, as a result, it reduces dimensionality of the magnetic interactions and frustration as well, as found in ZnV$_2$O$_4$ (36). Therefore, the spin glassy state of $Y_2Mo_2O_7$ may be due to spatially random coupling constants induced by the orbital degrees of...
freedom rather than strong frustration. The effectively reduced dimensionality may be related to the observed $C_s \propto T^{\gamma}$ with $D < 3$. A previous muon spin relaxation study (37) showed that the spin glassy state of Y$_2$MnO$_7$ seems to remain intact at 20% of non-magnetic doping as in SCGO, which may indicate that the jamming physics is still relevant to Y$_2$MnO$_7$ as well. We believe that the concept of jamming and complex energy landscape can unify the seemingly different glassy states found in dense magnetic systems, such as SCGO, Y$_2$MnO$_7$ (30), spin ice (38), and even the dynamics of magnetic domain boundaries (39).

Conclusion

The search for glassiness that arises intrinsically without defects and randomness has been revived recently as such glassiness may bear intricate relations with topological order (40), lack of thermalization in many body localization (41), jamming in structural glasses (42), and glassiness in supercooled liquids (43). Our experiments indicate that quantum fluctuations, via an order by fluctuations mechanism, induce a glassy state, a spin jam, in the strongly frustrated SCGO($p > 0.8$), which is robust, and extends to the clean limit. The findings strongly support the possible existence of purely topological glassy states.

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