

flux is conserved by all cyclic exchanges of spins on loops of alternating u and d spins, motivating a loop expansion of the fluctuation contribution to the free energy^{38–42}. The leading term in such an expansion counts the number N_{flip} of six-site hexagonal rings in a “flippable” $u-d-u-d-u-d$ configuration.

In Fig. 15(b) we plot the fluctuation entropy s_f as a function of N_{flip} . The highest (lowest) values are achieved for the 16-sublattice (four-sublattice) ordered states with the most (least) flippable hexagons. The fact that randomly generated $uuud$ states lie extremely close to the line connecting these two states suggests that loops of more than six sites contribute little to the fluctuation entropy.

The remaining question is why the overall difference in fluctuation entropy between different $uuud$ states is so small? We can express the fluctuation entropy in terms of the $2N$ eigenvalues $\{\varepsilon_n\}$ of \mathcal{M} as

$$s_f = -\frac{1}{2N} \sum_n \ln \varepsilon_n. \quad (41)$$

The eigenvalue spectrum $\{\varepsilon_n\}$ associated with the simplest $\mathbf{q} = 0$ four-sublattice $uuud$ state can easily be calculated analytically; working with a four-site unit cell, there are four bands, one of which is nondispersive. The associated density of states (DOS) for $b = 0.6$ is shown in Fig. 16(a), where the flat band appears as a sharp peak at $\varepsilon = 16b + h - 4$. In Figs. 16(b)–(d) we compare the integrated DOS from these four bands with numerical results for the integrated DOS of a 1024-site cluster.

The integrated DOS, averaged within the dimer manifold of $uuud$ states [Fig. 16(d)], is indistinguishable by eye from that of the four-sublattice state [Fig. 16(b)]. The step associated with the flat band survives as a set of $N/4$ localized excitations at $\varepsilon = 16b$. And, critically, the gap

$$\Delta = 8b + 4 - \sqrt{(8b + 4)(8b + 4 - h) + h^2} \quad (42)$$

to the lowest lying excitation is set by a *nodeless* eigenvector, whose components δS_i^α depend only on whether the spin \mathbf{S}_i points up or down. All $uuud$ states can be made formally equivalent to four-sublattice order by renumbering the sites in each individual tetrahedron, and the energy of this nodeless excitation is also unchanged by this renumbering of sites. It is therefore completely insensitive to whether or not the system is ordered. From the results it is clear why the thermodynamic properties of the plateau liquid state, and in particular the entropy associated with fluctuations about it, are so close to those of the ordered plateau state.

From these results, it is also possible to understand why the numerically determined entropy gain Δs_f increases as $b \rightarrow 0$ for $h = 4$ (cf. Table II). This singular behavior can be traced back to a band of excitations above the spin-wave gap $\Delta \approx 4b$, with bandwidth $\Delta\varepsilon \sim b$, which collapses to become a strict set of zero modes for $b \rightarrow 0$. Since zero modes are excluded from the sum which determines Δs_f , while the collapsing band contributes as $\sim \ln b$, b acts as a singular perturbation, and infinitesimal b may drive the system to order. This is despite the fact that it is disordered for $b = 0$, and for the relatively large b used in our simulations.

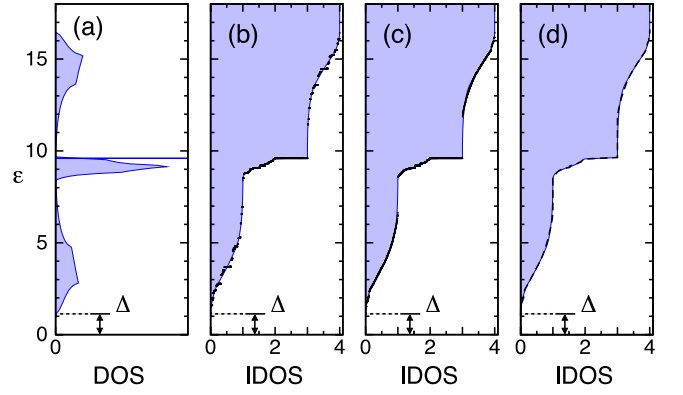


FIG. 16: (Color online) (a) Density of states (DOS) for eigenvalues of \mathcal{M} for four-sublattice $uuud$ state in thermodynamic limit, showing finite gap Δ [Eq. (42)] and flat band at $\varepsilon = 16b$. Integrated DOS (IDOS) (b) of four-sublattice $uuud$ state (points), (c) of a typical disordered $uuud$ state (points), and (d) averaged within disordered $uuud$ states (dashed line). In all cases $J_3 = 0$, $h = 4$, and $b = 0.6$, and a cluster size is $N = 1024$. The integrated DOS corresponding to (a) is shown in shading on (b)–(d) for comparison.

D. vector-multipole phase with local T_2 symmetry

At the upper critical field of the magnetization plateau, the collinear spins of the $uuud$ configurations cant away from the z axis. This instability occurs at the level of a single tetrahedron (Fig. 3), where it is continuous. On a lattice, it is associated with the closing of the gap Δ [Eq. (42)] in the excitation spectrum of the plateau liquid. Because of the special structure of this excitation, discussed above, the gap closes at the same value of $h_c = 4 + 8b$ for all $uuud$ states, and the transition is once again continuous — at least for $T = 0$. However, since the spin configurations in question are simply 3:1 canted versions of the $uuud$ states, with local T_2 symmetry, all of the entropic arguments presented above for the plateau liquid still hold. Thermal fluctuations alone cannot restore (canted) Néel order, and spin-spin correlations exhibit a power-law decay of $1/r^3$ for $T \rightarrow 0$.

The resulting state does however exhibit long range order in *both* the rank-two tensor order parameters $\mathbf{Q}^{\perp,1}$ and $\mathbf{Q}^{\perp,2}$ [Eqs. (22) and (23), and Table I]. The 3:1 canting of the $uuud$ spins selects a direction in the xy plane, and the primary order parameter is therefore the lower-symmetry irrep, $\mathbf{Q}^{\perp,1}$. The finite value of the nematic order parameter $\mathbf{Q}^{\perp,2}$ reflects the fact that this canting is coplanar. Since $\mathbf{Q}^{\perp,1}$ transforms like a vector under rotations about the z axis, we classify this state as a vector-multipole phase with local T_2 symmetry.

Within the framework of a Ginzburg-Landau theory, the contribution to the free energy from this pair of order parameters is

$$\begin{aligned} \mathcal{F} = & a_1 |\mathbf{Q}^{\perp,1}|^2 + a_2 |\mathbf{Q}^{\perp,2}|^2 \\ & + b_{12} \left(Q_1^{\perp,2} \left[(Q_1^{\perp,1})^2 - (Q_2^{\perp,1})^2 \right] + 2Q_2^{\perp,2} Q_1^{\perp,1} Q_2^{\perp,1} \right) \\ & + c_{11} |\mathbf{Q}^{\perp,1}|^4 + 2c_{12} |\mathbf{Q}^{\perp,1}|^2 |\mathbf{Q}^{\perp,2}|^2 + c_{22} |\mathbf{Q}^{\perp,2}|^4, \end{aligned} \quad (43)$$