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Emergent Separation of Valence Bond Regimes in $LiZn_2Mo_3O_8^1$

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LiZn₂Mo₃O₈ is a S = 1/2 triangular lattice antiferromagnet in which the basic magnetic building block is an Mo3O13 cluster rather than an individual ion. Rather than forming the 120° ordered magnetic state expected for a Heisenberg nearest neighbor triangular antiferromagnet, LiZn₂Mo₃O₈ instead favors a complex valence bond order across different energy and lengthscales: approximately two thirds of the spins form singlets at T ~ 100 K, while the remainder form valence bonds at lower temperature, as indicated by inelastic neutron scattering. No static magnetic order is detected by uSR down to T = 0.05 K. What is the origin of this spontaneous separation into different regimes of magnetic fluctuations? Recent experimental data, including the impact of hole doping, will be compared to different theoretical models that have been proposed for this behavior, including an emergent honeycomb lattice by octahedral rotations and partial charge ordering driven by extra intracluster degrees of freedom.

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