Enhanced-Entropy Phases in Geometrically Frustrated Pyrochlore Magnets

P. Timsina,[1] A. Chappa,[1] D. Alyones,[2,1] I. Vasiliev,[1] and L. Miao[1,+]
[1] *Department of Physics, New Mexico State University, Las Cruces, NM, USA*
[2] *Department of Chemical and Materials Engineering, New Mexico State University, Las Cruces, NM, USA*

Frustrated magnets host unconventional states stabilized by degeneracy and entropy, from spin ice [1] to quantum spin liquids [2] and pyrochlore oxides [3]. Pyrochlore iridates $R_2Ir_2O_7$ (R = Dy, Ho) provide a platform with tunable *d-f* exchange interactions and multiple frustrated phases [3,4]. In these systems, competing interactions suppress long-range order, yielding emergent quasiparticles such as magnetic monopoles [1].

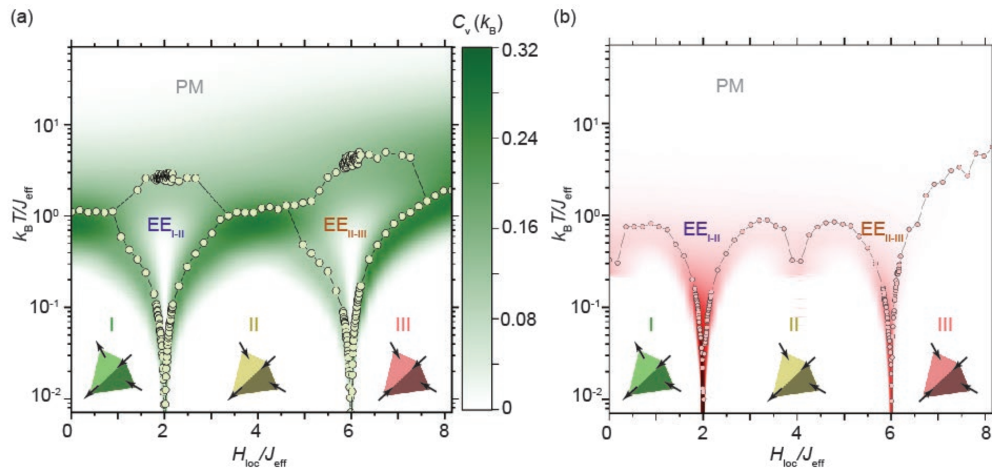

Figure 1: (a) Specific heat (C_v) and (b) susceptibility (χ) as a map of k_BT/J_{eff} and H_{loc}/J_{eff}, showing two enhanced-entropy (EE) phases (EE_{I-II} & EE_{II-III}) between spin-ice (I), fragmented (II), and AIAO phases (III).

Using Monte Carlo simulations, we map the thermodynamic phase diagram, identifying the 2-in–2-out (2I2O) spin ice, fragmented 3-in–1-out/1-in–3-out (3I1O/1I3O) [4], and all-in–all-out (AIAO) ground states [5]. In this talk, we will investigate the two finite-temperature enhanced-entropy (EE) phases near phase boundaries, characterized by high entropy, strong susceptibility, and mixed spin configurations. These phases are found to be stabilized by entropy-driven free-energy minimization, with distinct behavior of specific heat capacity decoupling from susceptibility serving as key signatures [5] (Fig. 1). These EE states define a new class of entropy-stabilized magnetic phases, underscoring the role of frustration in finite-temperature correlated states and offering pathways for entropy-based material design.

[1] A. P. Ramirez, A. Hayashi, R. J. Cava, R. Siddharthan, & B. S. Shastry, Nature **399**, 333 (1999).
[2] C. Broholm, R. J. Cava, S. A. Kivelson, D. G. Nocera, M. R. Norman, and T. Senthil, Science **367**, 263–273 (2020).
[3] J. S. Gardner, M. J. P. Gingras, and J. E. Greedan, Rev. Mod. Phys. **82**, 53 (2010).
[4] E. Lefrancois, V. Cathelin, E. Lhotel, J. Robert, P. Lejay, C.V. Colin, B. Canals, F. Damay, J. Ollivier, B. Fak, L. C. Chapon, R. Ballou, and V. Simonet, Nat. Commun. **8**, 209 (2017).
[5] P. Timsina, A. Chappa, D. Alyones, I. Vasiliev, and L. Miao, arXiv:2505.13352 (submitted: PRB, 2025).

+ Author for correspondence: lmiao@nmsu.edu

Suplementary information:

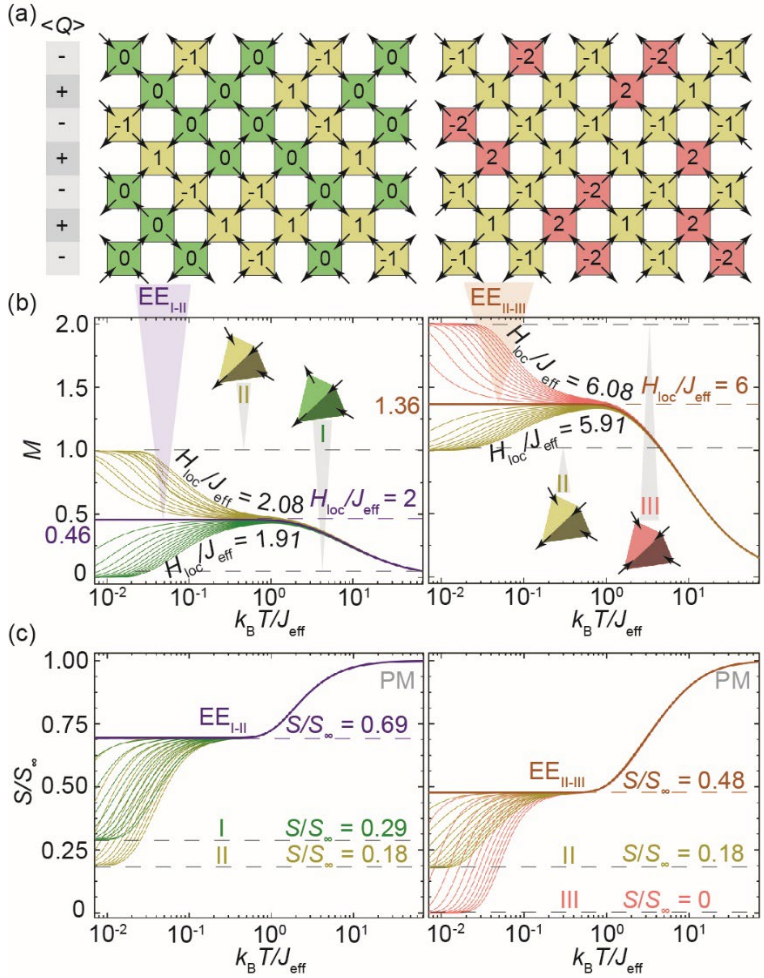

Figure 2: Spin configurations and entropy of EE phases. (a) Schematics of EE_{I-II} and EE_{I-II} phases with mixed tetrahedra fractions. (b) Magnetic order parameter (M) and (c) Normalized entropy (S/S_∞) versus k_BT/J_{eff}, confirming disordered and finite temperature stability.