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(G*) Constraints on the Spin Hamiltonian and Entropy of the Dipole-Octupole Spin Liquid Candidate Ce2Zr2O7 from Low Temperature Heat Capacity

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The Ce3+ pseudospin-1/2 degrees of freedom in the pyrochlore magnet Ce2Zr2O7 are known to possess dipole-octupole (DO) character, making it a candidate for novel quantum spin liquid (QSL) ground states at low temperatures. We report new heat capacity (CP) measurements on Ce2Zr2O7, which can be extrapolated to zero temperature to account for R·ln(2) entropy using a form appropriate to quantum spin ice. The measured CP rises sharply at low temperatures, initially plateauing near 0.08 K, before falling off towards a high temperature zero beyond 3 K. Phenomenologically, the entropy recovery above T = 0.08 K gives R·ln(2) less (R/2)·ln(3/2), the missing Pauling, spin ice entropy. At higher temperatures, the same data set can be fit to the results of a numerical linked cluster (NLC) calculation that allows estimates for the terms in the XYZ Hamiltonian expected for such DO pyrochlore systems. This constrains possible exotic and ordered ground states, and clearly favours the realization of a U(1)π QSL state. NLC calculations of the magnetic susceptibility and dynamic structure factor agree with these results and provide further constraints on the experimentally-determined values of the exchange parameters.

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