Supplementary Information for "Tensor network analysis of the maple-leaf antiferromagnet spangolite"

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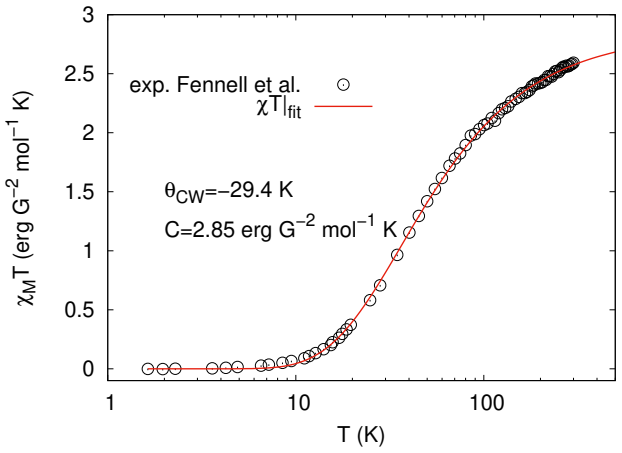
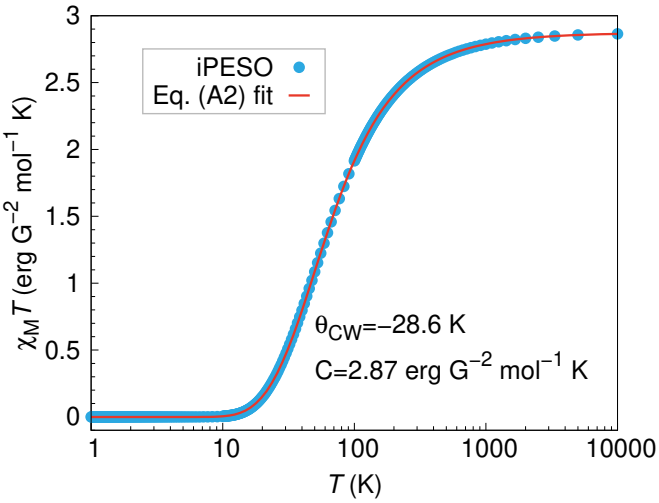
Supplementary Note 1. REVISION OF THE CURIE-WEISS TEMPERATURE

Supplementary Figure S1 shows the fit of the experimental magnetic susceptibility using the *ansatz* developed recently by Pohle and Jaubert [S1] as explained in the Method section. Supplementary Figure S2 is the consistency check, fitting the theoretical magnetic susceptibility using the same *ansatz*.



Supplementary Figure S1. Magnetic susceptibility $\chi(T)$ of spangolite from Ref. [S2], multiplied by temperature (symbols) and fit by the *ansatz* of Eq. 8 of the main paper.



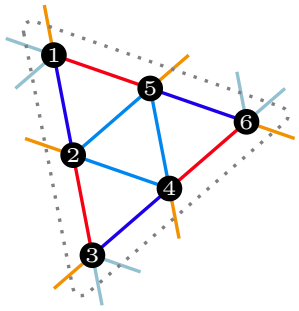
Supplementary Figure S2. Calculated magnetic susceptibility $\chi(T)$ of spangolite, multiplied by temperature (symbols) and fit by the *ansatz* of Eq. 8 of the main paper. The theoretical susceptibility data have been scaled by a factor of ~ 0.68 to match the high-temperature tail as explained in the main text.

Supplementary Note 2. MAPLE-LEAF LATTICE DEFINITION

Assuming a Cartesian coordinate system that sets the reference, the lattice vectors are given by

$$\mathbf{a}_1 = -\frac{\sqrt{7}}{2}\begin{pmatrix}1\\ \sqrt{3}\end{pmatrix} \qquad \mathbf{a}_2 = \sqrt{7}\begin{pmatrix}1\\ 0\end{pmatrix}. \tag{S1}$$

Here, the lattice constant is set to $a = 1$. Furthermore, the six-site basis of the maple-leaf lattice is defined in Supplementary Figure S3. A fully translational system therefore consists of a single unit cell with six spins. The basis is



Supplementary Figure S3. Definition of one unit cell, the six-site basis of the maple-leaf lattice.

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spanned by the six basis vectors

$$\begin{aligned} \mathbf{b}_1 &= \begin{pmatrix} 0 \\ 0 \end{pmatrix} & \mathbf{b}_4 &= \frac{1}{\sqrt{7}} \begin{pmatrix} 3 \\ -2\sqrt{3} \end{pmatrix} \\ \mathbf{b}_2 &= \frac{1}{2\sqrt{7}} \begin{pmatrix} 1 \\ -3\sqrt{3} \end{pmatrix} & \mathbf{b}_5 &= \frac{1}{2\sqrt{7}} \begin{pmatrix} 5 \\ -\sqrt{3} \end{pmatrix} \\ \mathbf{b}_3 &= \frac{1}{\sqrt{7}} \begin{pmatrix} 1 \\ -3\sqrt{3} \end{pmatrix} & \mathbf{b}_6 &= \frac{1}{\sqrt{7}} \begin{pmatrix} 5 \\ -\sqrt{3} \end{pmatrix}. \end{aligned} \tag{S2}$$

Supplementary Note 3. DETAILS OF THE TENSOR NETWORK CALCULATIONS

A. Ground state calculations

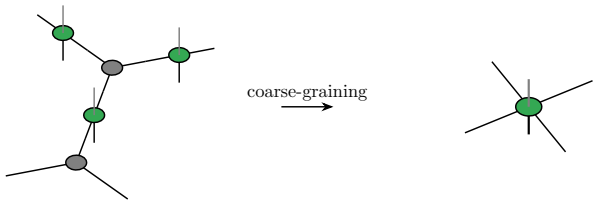
In order to represent the wave function of the ground state, we employ the iPESS ansatz. The tensor coefficients, that ultimately represent the target wave function in the thermodynamic limit are determined by a simple update procedure. It is based on evolving an initial wave function under imaginary time evolution to project out the ground state [S3, S4]. Due to the chosen coarse-graining of J_1 bonds, a single update step includes six lattice sites in triangle configurations at a time. This is visualized in Fig. 2 **b** of the main paper. In order to keep the bond dimension constant, the network has to be truncated back to χ_B after each application of the imaginary time evolution gate $\mathrm{e}^{-\delta\tau H}$. Although the simple update is an approximate scheme and treats the environment of a cluster only in mean-field approximation, it is expected and has proven to work reliably for frustrated quantum lattice systems in two dimensions.

B. Thermal state calculations

In contrast to ground state simulations, the local spin degrees of freedom have to be doubled, such that the system is described by a thermal density matrix. The iPESS ansatz for state vectors has been recently extended to the realm of operators, enabling the efficient simulation of thermal states of frustrated systems using iPESO [S5]. Here, the algorithm used to obtain a thermal density matrix $\rho(\beta)$ at inverse temperature β is again based on the simple update and Trotterization of the Hamiltonian. Starting from an infinite-temperature state $\rho(\beta = 0)$, where the thermal density matrix is simply a tensor product of local identity matrices, the system is cooled down to the desired final temperature β. This procedure is implemented by successive cooling steps with infinitesimal temperature $\delta\beta$ to keep the overall errors controllable. This can be done in a similar fashion as in the simple update ground state optimization and it is visualized in Fig. 2 **c** of the main paper. Again, after the evolution with the gate $\mathrm{e}^{-\delta\beta H}$, the resulting iPESO network is truncated to the bulk bond dimension χ_B. The accumulated truncation error ε of the full procedure can be used to probe the accuracy of the simulations [S5].

C. Calculation of expectation values

In order to compute expectation values, the honeycomb structure of the iPESS/iPESO ansatz can be mapped to a regular square lattice with lattice vectors as shown in Fig. 1 **a** of the main paper. This procedure is visualized in Supplementary Figure S4. The resulting tensor network on the square lattice is known as an *infinite projected entangled pair state* (iPEPS) or *infinite projected entangled pair operator* (iPEPO) respectively, which includes six spins of the original MLL on every lattice site. This additional step is required to compute effective fixed-point environments of the infinite MLL, e.g. by a regular *corner transfer matrix renormalization group* (CTMRG) procedure, as shown in Fig. 2 **d** of the main paper. Expectation values on the MLL can then be accurately computed by the evaluation of single-site and three-site expectation values on the coarse-grained square lattice, corresponding to the two different types of simplex configurations in the iPESS/iPESO ansatz. The approximations in the contraction of the infinite 2D lattice are controlled by a bond dimension for the effective environment tensors, denoted by χ_E. In practise, this parameter has to be large enough so that expectation values are well converged. Given the strong dimerisation observed for spangolite, the choice of $\chi_\mathrm{E} = \chi_\mathrm{B}^2$ was found to be sufficiently accurate. For $\chi_\mathrm{B} > 8$, we fixed $\chi_\mathrm{E} = \chi_\mathrm{B}\,(\chi_\mathrm{B} - 2)$. Besides taking the full CTMRG environment into account for the calculation of expectation values, it is also possible to only use a mean-field (MF) environment. This environment is readily available from the simple update for both ground and thermal states. MF expectation values are computationally much cheaper and have to be used for large bond dimensions due to the high computational cost of the CTMRG routine.



Supplementary Figure S4. Coarse-graining of the three-site unit cell to map the iPESS/iPESO tensor network ansatz on the honeycomb lattice to a regular square lattice. Each lattice site then contains six spins of the original maple-leaf lattice.

SUPPLEMENTARY REFERENCES

[S1] R. Pohle and L. D. C. Jaubert, Curie-law crossover in spin liquids, Phys. Rev. B **108**, 024411 (2023).

[S2] T. Fennell, J. O. Piatek, R. A. Stephenson, G. J. Nilsen, and H. M. Rønnow, Spangolite: an s = 1/2 maple leaf lattice antiferromagnet?, J. Phys.: Condens. Matter **23**, 164201 (2011).

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