A quantum magnetic analogue to the critical point of water

At the liquid–gas phase transition in water, the density has a discontinuity at atmospheric pressure; however, the line of these first-order transitions defined by increasing the applied pressure terminates at the critical point, a concept ubiquitous in statistical thermodynamics. In correlated quantum materials, it was predicted and then confirmed experimentally that a critical point terminates the line of Mott metal–insulator transitions, which are also first-order with a discontinuous charge carrier density. In quantum spin systems, continuous quantum phase transitions have been controlled by pressure, applied magnetic field and disorder, but discontinuous quantum phase transitions have received less attention. The geometrically frustrated quantum antiferromagnet \( \text{SrCu}_2(\text{BO}_3)_2 \) constitutes a near-exact realization of the paradigmatic Shastry–Sutherland model and displays exotic phenomena including magnetization plateaus, low-lying bound-state excitations, anomalous thermodynamics and discontinuous quantum phase transitions. Here we control both the pressure and the magnetic field applied to \( \text{SrCu}_2(\text{BO}_3)_2 \) to provide evidence of critical-point physics in a pure spin system. We use high-precision specific-heat measurements to demonstrate that, as in water, the pressure–temperature phase diagram has a first-order transition line that separates phases with different local magnetic energy densities, and that terminates at an Ising critical point. We provide a quantitative explanation of our data using recently developed finite-temperature tensor-network methods. These results further our understanding of first-order quantum phase transitions in quantum magnetism, with potential applications in materials where anisotropic spin interactions produce the topological properties that are useful for spintronic applications.

In the pressure–temperature \((P,T)\) phase diagram of water (Fig. 1a), the line of first-order transitions terminates at \(P_c = 221\) bar and \(T_c = 374\) °C, defining the critical point where liquid and vapour become a single phase. Although the first-order line has no critical scaling properties, its termination point does. In the supercritical regime, around and above the critical point, there is no transition and indeed one may proceed continuously from liquid to vapour without ever undergoing one. Because their difference is defined not by a change in symmetry but by a scalar, the particle density, which can take two values at the discontinuity, the critical point is in the Ising universality class.

Quantum systems with only spin degrees of freedom enable theorists to realize entangled quantum many-body models, including those with exact solutions or ground states, and experimentalists to probe their properties on truly macroscopic lengthscales. An example of a two-dimensional (2D) quantum spin system that hosts a first-order quantum phase transition (QPT) between two exactly known ground states is provided by the \( S = 1/2 \) Heisenberg model in a fully frustrated bilayer geometry. In that system, it was shown theoretically that the line of first-order transitions extending to finite temperatures is again terminated by an Ising critical point, as we summarize in the Methods section.

A quantum magnetic material with a related geometry, \( \text{SrCu}_2(\text{BO}_3)_2 \) (Ref. 28), has drawn intensive interest because of the extreme frustration of its orthogonal-dimer structure (Fig. 1b, inset). This realizes a \( S = 1/2 \) Heisenberg model formulated by Shastry and Sutherland because of its exact ground state, a product of dimer singlets, at all small and intermediate values of the inter- to intra-dimer interaction ratios (\( J_{\text{dd}} < 0.675 \)). Although the frustration is manifest in many unusual...
phenomenal, our interest in the Shastry–Sutherland model lies first in the presence of two first-order QPTs, from the dimer phase to a plaquette phase at $J/J_0 = 0.675(2)$ and thence to an ordered Néel antiferromagnet at $J/J_0 = 0.765(1)$, and second in the fact that applying a hydrostatic pressure to SrCu$_2$(BO$_3$)$_2$ acts to control $J/J_0$, revealing both transitions at respective pressures of approximately 19 kbar (ref. 18) and 27 kbar (ref. 19).

To investigate the possibility of a critical point in SrCu$_2$(BO$_3$)$_2$, we perform high-precision measurements of the specific heat using an a.c. calorimetry technique. Large single crystals of SrCu$_2$(BO$_3$)$_2$ were grown by a floating-zone method (Methods). Samples of masses up to 36 mg were cut, patterned with metallic strips for calorimetry and mounted in a clamp cell allowing hydrostatic pressures up to 26.5 kbar and in applied magnetic fields up to 9 T. Details of our a.c. measurement procedures are provided in Methods and Extended Data Fig. 1.

**Results: zero field**

Starting at zero magnetic field, the pressure-induced evolution of the specific heat, shown as $C(T)/T$, is illustrated in Fig. 1b. As quantified in Fig. 2a, $C(T)/T$ at low pressures shows an exponential rise to a broad maximum at a temperature, $T_{\text{max}}$, that tracks the gap to the triplon or bound-triplon excitations of the dimer phase. With increasing $P$, this peak moves gradually lower and becomes proportionally narrower, but between 18 and 20 kbar it becomes extremely tall and narrow (Figs. 1b, 2a), bearing all the characteristics of a critical point. After reaching a lowest measured value of $3.4 K$ at $P = 20$ kbar, $T_{\text{max}}$ rises with increasing pressure and the peak broadens again (Fig. 2b), indicating that the singular behaviour has terminated. A second small peak appears around 2 K for $P > 18$ kbar (Figs. 1b, 2a, b) and persists to our upper pressure limit. We expect that this feature marks the thermal transition out of the plaquette phase which in SrCu$_2$(BO$_3$)$_2$ was shown to be an approximately linear function of the applied pressure. Correlation length, $\xi$, obtained by iPEPS with $D = 20$ and expressed in units of the lattice constant, $a$, becomes large only at the finite-temperature critical point; the dashed black line shows the locus of maxima of $\xi(J/J_0)$ at each fixed temperature, and terminates when $J/a < 1$. We note that the colour scales in c and d are truncated such that they do not include the peak values of $C/T$ and $\xi$. D. Dimer spin–spin correlation function, $\langle S_i S_j \rangle$, showing a discontinuity with $J/J_0$ at low temperatures but continuous behaviour throughout the supercritical regime. The dashed black line, the equivalent of the critical isochore in water, shows the locus of points where this order parameter is constant at its critical-point value, $\langle S_i S_j \rangle = -0.372(30)$.
Fig. 2 | Specific heat and critical scaling at zero magnetic field. a, b, $C(T)/T$ measured for applied pressures from 0 to 18.2 kbar (a) and from 20 to 26.5 kbar (b). The temperature of the broad peak that is observed at most pressures ($P \leq 15$ kbar and $P \geq 22$ kbar) drops to a finite minimum value at the QPT, where $C(T)/T$ displays only a tall and extremely narrow peak. c, d, Specific heat obtained by $D = 20$ iPEPS calculations performed for the Shastry–Sutherland model in the dimer (c) and plaquette (d) phases using plaquette-based tensors (Methods). The evolution of peak heights with proximity to the critical coupling ratio, illustrated clearly in the numerical data, is less apparent in experiment. We note that the phonon contribution ($C(T)/T \approx C(T)/T^2$) to the measured specific heat becomes appreciable at higher temperatures; although this can be subtracted for accurate fitting	extsuperscript{23}, our focus here is on the peak positions at and below 6 K. e, Convergence of the critical coupling ratio obtained in finite-temperature iPEPS calculations as a function of $1/D$ (solid circles); the extrapolated value of 0.671 (open square) agrees well with the zero-temperature value	extsuperscript{10, 11} f, Convergence of the critical temperature as a function of $1/D$ (solid circles), leading to the estimate $k_B T_c/J_0 = 0.039(6)$ (open square). The error bars on the extrapolated values (open squares) are conservative estimates based on their separation from the last calculated point. g, Critical exponent, $\beta = 0.123(15)$, of the discontinuity, $\Delta(S\cdot S)$, in the dimer spin–spin correlation function at the first-order transition line, demonstrating consistency with the 2D Ising exponent, $\beta = 1/8$. This estimate is consistent with previous theoretical calculations	extsuperscript{8},	extsuperscript{11}.
identified an isolated critical point (Fig. 1e), with the Ising exponent $\beta = 1/8$ (Fig. 2g), in the Shastry–Sutherland model. Thus our results confirm scenario (i) and we place the critical point of SrCu$_2$(BO$_3$)$_2$ at $P_c = 19$ kbar and $T_c = 3.3$ K.

**Results in a magnetic field**

To challenge our interpretation, we consider the situation in a finite magnetic field. Because the field has little effect on the gapped dimer and plaquette phases, the physics of the critical point should be essentially unaffected. We have measured the specific heat in fields up to $\mu_0 H = 9$ T ($\mu_0$, vacuum permeability; $H$, magnetic field strength), and indeed we observe near the critical point (18.2 kbar, Fig. 3a) that the peak remains sharp and shows only minor field-induced changes. These features are reproduced in detail by our iPEPS results, shown in Fig. 3d for $h/H_0$ value very close to the $D = 14$ QPT; the results also indicate that the small changes can result simply from not pinpointing the exact critical coupling.

Turning to the peak around 2 K, our 18.2-kbar and 22.8-kbar data (Fig. 3a, b) show that the field suppresses both its height and position. However, above 5.5 T for 22.8 kbar, another peak has emerged that is very different in shape, becoming tall, sharp and isolated from the broad 4-K hump, which has almost vanished above 4 T in our 26.5-kbar data (Fig. 3c). In Fig. 3e we collect all of these peaks to obtain a clear picture of three key phenomena. First, the 4-K features remain isolated, changing from sharp to broad with increasing $P$ and $H$ in a manner mostly independent of the complex action below 2.5 K. Second, the thermal transition of the plaquette phase is suppressed to zero temperature by fields of 6 T at 18.2 and 20.0 kbar; this field value is consistent with a closing of the gap to the triplet plaquette excitation observed in previous work. Third, this suppression is replaced at our higher pressures and fields by the emergence at finite temperatures of the new, sharp peak, which from its field-induced evolution we attribute to the ordering transition of the antiferromagnetic phase.

We note that zero-field $C(T)$ data similar to ours have appeared recently. These authors did observe the tall, sharp critical-point peak in their data at 19 and 21 kbar (1.9 and 2.1 GPa, shown only in their supplemental materials), but provided no explanation. They also observed the 2-K peak and ascribed it to a plaquette state (which they assume to be the ‘empty’ plaquette phase, see below), the deduced gap of which is also consistent with the 6-T critical field we observe. At pressures just beyond the limits of our study, they also argued that the pressure-induced development of the 2-K feature is a signature of the antiferromagnetic phase, and hence our observation of strong,

![Fig. 3 | Evolution of the specific heat with magnetic field around the QPT.](image-url)
sharp features in an applied field (Fig. 3b, c) helps to confirm their identification.

Phase diagram

In Fig. 4 we gather all the features abstracted from our specific-heat data as a function of both the applied magnetic field and the pressure. The $(P, T)$ plane shows the critical point (red star) lying well above the line of thermal Ising transitions. On the scale of the figure, the first-order line is essentially vertical in temperature. Although a structural transition is probably associated with the magnetic transition in SrCu$_2$(BO$_3$)$_2$, causing a discontinuity in the coupling constants, we stress that there is only one transition as a function of $|J|/J_0$, (Fig. 1). This picture of the critical point at $H = 0$ is reinforced by the data at finite fields, which indicate a line of critical points, and a wall of first-order transitions, that dominate the $(P, H, T)$ phase diagram. Below $T_c(P, H)$, this wall defines a plane through which the average dimer spin–spin correlation should change discontinuously, a prediction that may be tested by measuring the $P$ dependence of the instantaneous spin correlation function by neutron scattering, or of phonon modes sensitive to the magnetic correlations.

At temperatures below the critical point in Fig. 4, our data reveal a wealth of quantum and thermal phase transitions. The field suppresses the continuous thermal transitions out of the plaquette phase and in its place we have found the antiferromagnetic phase at pressures as low as 22.8 kbar. The pressure-induced plaquette–antiferromagnetic QPT was thought to be weakly first-order, but has recently been proposed as a candidate deconfined quantum critical point. Although our data suggest that the field-induced plaquette–antiferromagnetic QPT could change from first to second order between different regimes, there is no clear sign of another finite-temperature critical point. Establishing the universality classes of these transitions remains a challenge that is driving progress both in specific-heat measurements under extreme $(P, H, T)$ conditions and in numerical methods for computing the thermal properties of frustrated systems.

Although our iPEPS studies of the Shastry–Sutherland model give a quantitative account of the critical-point physics of SrCu$_2$(BO$_3$)$_2$, they differ from experimental data on the nature of the plaquette–ordered phase. NMR and neutron scattering experiments indicate that in the plaquette phase of SrCu$_2$(BO$_3$)$_2$, the singlets form on the ‘full’ plaquettes of the $J_l$ lattice (those with a $J_l$ bond in the inset of Fig. 1b), whereas the Shastry–Sutherland model favours singlets on the ‘empty’ plaquettes. From the sensitive energetic competition between the empty-plaquette and full-plaquette phases, it is not surprising that the additional three-dimensional and Dzyaloshinskii–Moriya terms in SrCu$_2$(BO$_3$)$_2$ could cause this discrepancy, and we expect the state we observe above 18 kbar and below 2 K to be the full-plaquette phase. Because both plaquette phases have twofold degenerate ground states (only half of the plaquettes may form singlets), the thermal transition has Ising symmetry independent of their empty-plaquette or full-plaquette nature, and our results are not affected by this issue.

The physics of the critical-point phase diagram is different from that of second-order QPTs: because one may pass from one side of the discontinuity to the other without crossing a transition, there is no breaking of symmetry. At the critical point itself, the property of a divergent correlation length (Fig. 1d, Extended Data Fig. 2), associated with domain sizes and having critical exponents set by the universality class, remains. The supercritical regime has recently become a subject of active investigation, including (with a view to sensitive switching) in critical fluids. In the Mott metal–insulator phase diagram, and its realization in the Hubbard model, the striking feature of the phase diagram is that the temperature, $T_{\text{max}}$, characterizing the peak in $C/T$ reaches a minimum at $T_c$. We show in Extended Data Fig. 4 that this behaviour is universal around an Ising critical point in 2D lattice models. Thus the specific heat clearly defines not one but two characteristic lines in the supercritical regime (Fig. 1b, c), in contrast to the correlation length (Fig. 1d) and the critical isochore (Fig. 1e), both of which are regarded as marking a single crossover line.
Although this remarkable property of the specific heat is different from that in water (Fig. 1a), we stress that it is intrinsic to a system as simple as the Ising model. While the origin of this complex critical point physics awaits further theoretical analysis, we observe that its consequences can be probed experimentally in quantum spin systems by comparing the specific heat with scattering measurements of the order parameter. As modern quantum magnetism and spintronics embrace the highly spin-anisotropic interactions required to produce the topological physics of Ising, Kitaev, skyrmion and other systems, a full understanding of the resulting first-order QPTs will include the quantum phenomenology of the critical point, the classical variant of which has been known to science for two centuries.

Online content
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Article

Methods

Samples
Single crystals of SrCu₂(BO₃)₂ were grown by a floating-zone method. First, polycrystalline SrCu₂(BO₃)₂ was prepared by a solid-state reaction using as starting materials SrCO₃, CuO and B₂O₃ with 99.99% purity. These were mixed, ground and heat-treated at 900 °C in flowing oxygen for over 100 h with several intermediate grindings. The phase purity was verified by conventional X-ray diffractometry and the powder was pressed hydrostatically into rods (8 mm in diameter and around 90 mm in length) which were sintered at 900 °C for 24 h in an oxygen atmosphere.

Single-crystal growth was carried out using an optical floating zone furnace (FZ-T-10000-H-IV-VP-PC, Crystal Systems) with four 300-W halogen lamps as the heat source. Although the first reported single crystals of SrCu₂(BO₃)₂ were also grown in this type of furnace, using LiBO₂ discs as a solvent placed on a top of a seed rod, we obtained better results using a self-adjusted flux method. Optimal growth conditions were found to be a steady growth rate of 0.25 mm h⁻¹ at all times, rotation of both feeding and seeding rods at approximately 15 rpm in opposite directions to ensure homogeneity of the liquid and the application of a 5-bar pressure of argon with 20% oxygen. When a homogeneous melt was achieved, the power of the lamps was decreased slowly (over 24 h) until steady-state conditions were found (approximately 48.5% lamp power in the present case), after which these conditions were maintained rigorously until the end of the growth. The full growth process required approximately two weeks and resulted in a recrystallized boule 8 cm in length containing a single-crystalline grain in its top 4 cm.

Specific-heat measurements
Three slabs were cut from the single-crystal rod, each with a cross-section of 7.6–9.0 mm² in the ab plane and a thickness of 0.5–1.0 mm parallel to the c axis. The sample masses were measured to be between 15 and 36 mg. The samples were polished gently to deposit Pt films of 20-nm thickness. To measure the heat capacity, C(P, H, T), under multiple extreme conditions (very low temperatures, high pressures and intense magnetic fields), we used a.c. calorimetry at a second-harmonic mode (2ω). We employed a piston cylinder BeCu clamp cell to apply pressures up to 26.5 kbar, with liquid kerosene as the pressure-transmitting medium. Measurements of the electrical resistivity at the superconducting transition of a Pb strip were used as a pressure manometer and revealed good hydrostatic conditions. The sample masses were measured to an accuracy of 5% with respect to an astatic technique. Further details concerning all aspects of the procedures of our a.c. calorimetry measurements may be found in previous work.

Fully frustrated bilayer
The Heisenberg model on the fully frustrated bilayer presents a clear, and thus conceptually valuable, example of a first-order QPT giving rise to a critical point at finite temperature. In this geometry, which has no known materials analogue, spin pairs (with coupling J) are arranged vertically on a square lattice with equal couplings (J) to both spins of all four dimer neighbours, and the ground state jumps discontinuously from exact dimer singlets to exact triplets at J/J₀ = 2.315. Increasing temperature, the discontinuity in triplet density reduces until the line of first-order transitions terminates at a critical point, in the 2D Ising universality class, with k_B T_c = 0.52J (ref. 29). The geometry of the fully frustrated bilayer has a direct connection to that of the Shastry–Sutherland model,27 suggesting that the two may share similar critical-point physics. A discontinuity in the average dimer spin–spin correlation is certainly well documented at the dimer–quadruplet QPT in the Shastry–Sutherland model. However, the models do differ in that the total spin of each dimer is a good quantum number in the fully frustrated bilayer, but not in the Shastry–Sutherland case. A further point of contrast is that the QPT of the fully frustrated bilayer is from the dimer–singlet phase to a 2D antiferromagnetic phase, which by the Mermin–Wagner theorem has no finite-temperature transition, forcing the critical point associated with the discontinuity to be observed as an isolated point. The fact that the plaquette phase of the Shastry–Sutherland model has a thermal Ising transition raises in principle the alternative scenario of the tricritical point, which we show is excluded by our experimental and numerical results.

iPEPS
Tensor-network methods provide a powerful approach for accurate numerical calculations of both the ground and thermal states of gapped local Hamiltonians. iPEPS,20,21,46, a 2D generalization of matrix-product states, are a variational ansatz enabling both wavefunctions and thermal states to be represented efficiently in the thermodynamic limit, with the accuracy of the representation controlled systematically by the bond dimension, D, of the tensors.

To obtain an iPEPS representation of a thermal state, we employ the algorithms used previously,23 which are based on the imaginary-time...
evolution of a purification of the thermal density operator. At each time step the bond dimension of the iPEPS is truncated to the maximal $D$. To maximize $D$ while working near the QPT, we restrict the truncation to the (local) simple-update approach. To improve the efficiency of the calculations, we exploit the global $U(1)$ symmetry of the model, which for a pure Heisenberg model is preserved in the presence of an applied field at finite temperatures. Physical expectation values are computed by contracting the tensor network, which we perform by a development of the corner-transfer-matrix method. This method is also used to compute the transfer matrix, from which we extract the correlation length, $\xi$, in Fig. 1d and Extended Data Fig. 3b, of the correlation function with the slowest decay. iPEPS with a sufficiently large but finite $D$ can be used to represent a critical state at finite temperature, as in the early example of the 2D Ising model. In the Shastry–Sutherland model we have found that $D=8$ is already large enough to obtain a diverging $\xi$ on approaching the critical point, as we show in Extended Data Fig. 2; we remark that the region of parameter space over which the divergence is visible is considerably narrower than the point spacing of 0.001 shown in Fig. 1 and Extended Data Fig. 3.

In the Shastry–Sutherland problem, the optimal tensor geometry depends on the ground state; the spin correlations in the plaquette phase are represented most efficiently using one tensor per dimer in a two-dimer unit cell, whereas correlations in the plaquette phase are best described by an ansatz with one tensor per four sites (from four different dimers) on a plaquette. For working around a critical point, it is essential to use a single representation of both quantum phases, and in the Shastry–Sutherland model the plaquette basis is more efficient for representing the dimer phase than the reverse. For this reason, we have used the plaquette basis for all of the results reported here.

As a consequence we do observe numerical instabilities at the lowest temperatures when working in the dimer phase (the regions excluded from Figs. 1c–e, 2c, d). These are most probably an artefact arising because of the product nature of the dimer ground state, as all observables and energies are essentially converged at those temperatures. One means of circumventing this problem is to add a small Dzyaloshinskii–Moriya interaction to the model, in fact guided by the real spin Hamiltonian of SrCu$_3$(BO$_2$)$_2$. This leads to an entangled ground state, which, as shown in Extended Data Fig. 3, makes the low-temperature regime accessible, albeit at cost of a reduced $D$. These results confirm further that the Dzyaloshinskii–Moriya interactions of SrCu$_3$(BO$_2$)$_2$ have no effect on the physics of the critical point.

When working in the plaquette phase, the strongly entangled nature of the plaquette ground state makes low-temperature studies very challenging, and for this reason our primary focus is on temperatures $T/J_0 \gtrsim 0.03$, for a full characterization of the regime relevant to the critical point. Although a small feature is evident in $C/T$ around $T/J_0 = 0.02$ (Extended Data Fig. 3a), we caution against associating this with the Ising transition of the plaquette phase, because we do not find corresponding behaviour in the spin correlations and thus cannot confirm the reliability of our results at such low temperatures.

Data availability
The data that support the findings of this study are available at https://doi.org/10.5281/zenodo.4445613 and from the corresponding author upon reasonable request.

Code availability
The code that supports the findings of this study is available from the corresponding author upon reasonable request.

Additional information
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Correspondence and requests for materials should be addressed to H.M.R.

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Extended Data Fig. 1 | a.c. calorimetry on SrCu$_2$(BO$_3$)$_2$. a, The a.c. calorimeter was prepared by depositing two Pt thin films (the shinier surfaces) over both halves of the sample. One film was used as the heater and the other for optimal thermal contact and measurement. The heating current ($I_{ex}$ at frequency $f$) was supplied through the pair of Constantan wires labelled $H_{1a}$ and $H_{1b}$, and $H_{2a}$ and $H_{2b}$ were used to measure the electrical resistance, $R_{Pt}$, of the Pt film. TC$_1$ and TC$_2$ are thermocouples and $K$ represents the thermal contact between the sample and the cryostat (through the pressure cell). b, Isothermal ($T = 4.5$ K) and isobaric ($P = 20$ kbar) $f$-dependence of the modulated pick-up voltage, $V_{ac}$, which is directly proportional to the temperature differential, $\Delta T_{ac}$, measured by the thermocouples at two different positions. c, Isobaric ($P = 18.2$ kbar) $f$-dependence measurements of $V_{ac}$ at different temperatures with $I_0 = 1.6$ mA at $T \geq 3.9$ K, $I_0 = 0.8$ mA at $T = 2$ K and $I_0 = 0.4$ mA at $T < 2$ K. d, Sample heat capacity normalized to the input heating power ($P_{IR} = P_{t0}^2$), comparing the fit of $V_{ac}(f)$ obtained from the steady-state equation (‘f-scan’) with values obtained directly from a variable-temperature measurement performed at the fixed working frequency $f_C = 1.5$ Hz (‘T-scan’). Solid and dashed lines in b and c represent fits using the steady-state equation. 

Note: The diagram shows a calorimeter setup with labeled parts and measurement variables. The text describes the calorimetry setup and presents data for $V_{ac}$ and heat capacity under different conditions.
Extended Data Fig. 2 | Correlation length. $\xi/a$ for the Shastry–Sutherland model, calculated by iPEPS with $D = 8$ as a function of the coupling ratio, $J/J_0$, at a fixed temperature $T_c(D = 8) = 0.0522J_0/k_B$. The three panels show increasing magnification of the $J/J_0$ axis from the equivalent of Fig. 1 (upper right) through the step sizes of Fig. 2c, d ($10^{-3}J/J_0$, centre) to $10^{-5}J/J_0$ (lower left).
Extended Data Fig. 3 | Critical point in the presence of Dzyaloshinskii–Moriya interactions. Thermodynamic data obtained from iPEPS calculations with $D = 10$ performed for the Shastry–Sutherland model in the presence of Dzyaloshinskii–Moriya interactions. These interactions, of strength $D$, are placed on the dimer $(J_a)$ bonds and have the magnitude known for $\text{SrCu}_2(\text{BO}_3)_2$. They create an entangled ground state in the dimer phase, which resolves the numerical instabilities observed for the pure Shastry–Sutherland model at low temperatures, although the reduced symmetry limits the maximum $D$ to 10.

a, Specific heat, $C(J/J_D, T)/T$, shown in the same format as for Fig. 1b, c.
b, Correlation length, $\xi/a$, showing clearly the region of ‘pressure’ and temperature over which Ising correlations develop.
c, Dimer spin–spin correlation function, $\langle S_i \cdot S_j \rangle$, emphasizing the abrupt onset with decreasing temperature of a sharp discontinuity as a function of $J/J_D$. It is clear that these Dzyaloshinskii–Moriya interactions have no qualitative effect whatsoever on the physics of the critical point.
Extended Data Fig. 4 | Ising critical points in different lattice models.

Specific heat, $C/T$, for a number of 2D models, illustrating its universal behaviour around the Ising critical point. a, Ising model on the square lattice in a longitudinal magnetic field, $h$, obtained by contracting the exact $D = 2$ tensor-network representation of the partition function using the corner-transfer-matrix method with a boundary bond dimension $\chi = 24$ (ref. 55). b, Fully frustrated bilayer model, obtained by using the stochastic series expansion quantum Monte Carlo approach developed in refs. 27,45 to perform simulations on systems of sizes up to $2 \times 32 \times 32$ as a function of $J_{\perp}/J_{\parallel}$. c, Shastry–Sutherland model, obtained by iPEPS calculations with $D = 20$ as in Fig. 1c. The dashed lines show the positions of the local maxima of the specific heat, $C(J_{\perp}/h)$, which we label by their temperatures, $T_{\text{max}}$. These two lines reach an absolute minimum, $T_{\text{max}} = T_c$, where they meet at the Ising critical point, with $T_{\text{max}}$ increasing as the control parameter is changed away from the QPT. Thus the specific heat defines two characteristic lines in the phase diagram of the Ising critical point, instead of the single line given by the correlation length (Fig. 1d) and the critical isochore (Fig. 1e). This contrasting behaviour has been demonstrated in models where the critical pressure is temperature-independent and the issue of characteristic lines has also been discussed in the Mott metal–insulator phase diagram35,40. We stress that such behaviour is a fundamental property of the Ising model, and hence of all models sharing its physics. For the Shastry–Sutherland model (c), the two lines of maxima can be taken to provide a qualitative definition of regimes dominated by ‘dimer-like’ spin correlations (lower left) and by ‘plaquette-like’ correlations (lower right, but above the plaquette-ordered phase), accompanied by a third regime (above both lines) bearing no clear hallmarks of either $T = 0$ phase. We remark that the values of $T_c$ in units of the relevant energy scale, $T_c/J = 2.3$ (a), $T_c/J = 0.53$ (b) and $T_c/J = 0.04$ (c), vary widely among the three models. This can be traced to the change in slope of the ground-state energy at the transition, the compensation of which by entropy effects restores a derivable free energy at $T_c$. 

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**Notes:**
- **Extended Data Fig. 4** illustrates the specific heat, $C/T$, for a number of 2D models, including the Ising model, a fully frustrated bilayer model, and the Shastry–Sutherland model.
- The specific heat is shown as a function of various control parameters for each model, such as the longitudinal magnetic field $h$ for the Ising model and the ratio $J_{\perp}/J_{\parallel}$ for the fully frustrated bilayer model.
- The Ising model is obtained by contracting the exact $D = 2$ tensor-network representation using the corner-transfer-matrix method.
- The fully frustrated bilayer model is obtained using the stochastic series expansion quantum Monte Carlo approach.
- The Shastry–Sutherland model is obtained by iPEPS calculations.
- The dashed lines indicate the positions of the local maxima of the specific heat, which are labeled by their temperatures, $T_{\text{max}}$.
- These maxima reach an absolute minimum, $T_{\text{max}} = T_c$, at the Ising critical point.
- The specific heat defines two characteristic lines in the phase diagram of the Ising critical point.
- The values of $T_c$ in units of the relevant energy scale vary widely among the three models.
- The compensation of entropy effects plays a role in restoring a derivable free energy at $T_c$. 

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