Towards an experimental test of deconfined quantum criticality

Proximate deconfined quantum critical point in $SrCu_2(BO_3)_2$
Authors: Yi Cui, Lu Liu, Huihang Lin, Kai-Hsin Wu, Wenshan Hong, Xuefei Liu, Cong Li, Ze Hu, Ning Xi, Shiliang Li, Rong Yu, Anders W. Sandvik, Weiqiang Yu
Science 380, 1179 (2023)

Recommended with a Commentary by Frédéric Mila, Ecole Polytechnique Fédérale de Lausanne, Switzerland

According to Landau paradigm, quantum phase transitions that occur between two phases at zero temperature can be continuous, as their thermal counterparts, as long as one phase corresponds to breaking one symmetry (or more) with respect to the other phase. When the two phases break different symmetries, the transition should be first order. This general expectation has been challenged in 2004 by Senthil et al[1], who suggested an alternative scenario known as a Deconfined Quantum Critical Point (DQCP). According to this scenario, one phase is characterized by fractional excitations that are confined, i.e. that cannot exist without being in the proximity of one or more other fractional excitations, while the other phase is characterized by the development of a finite density of independent fractional excitations, a phenomenon also known as condensation. The transition corresponds to the deconfinement of the fractional excitations, hence the name. A typical example would be a valence bond crystal where spins are bound in singlets. Isolated spins can only exist at the meeting point of domain walls that separate different phases, and their deconfinement accompanied by condensation would lead to antiferromagnetic order. The symmetry broken in the valence bond crystal is a lattice symmetry while the antiferromagnetic phase breaks SU(2) symmetry in spin space. The two broken symmetries are unrelated, so the transition should be first order according to Landau theory, yet the transition could, according to Senthil et al, take place through a DQCP.

This prediction is now almost 20 years old, yet it has proven difficult to definitely check it in realistic models. The best candidate so far is a spin-1/2 model proposed by Sandvik[2] and known as the $J - Q$ model. In this model, the competition between the Heisenberg exchange term controlled by J and a four-spin term controlled by Q can be studied by Quantum Monte Carlo, and, according to these simulations, the transition between Néel order and columnar dimer order seems to be continuous. The nature of this putative critical point has been further investigated by field theory approaches[3], with the conclusion that this transition might actually be an exotic, weakly first-order one. In any case, scaling must be anomalous, suggesting that this critical point might be a multicritical point[4].

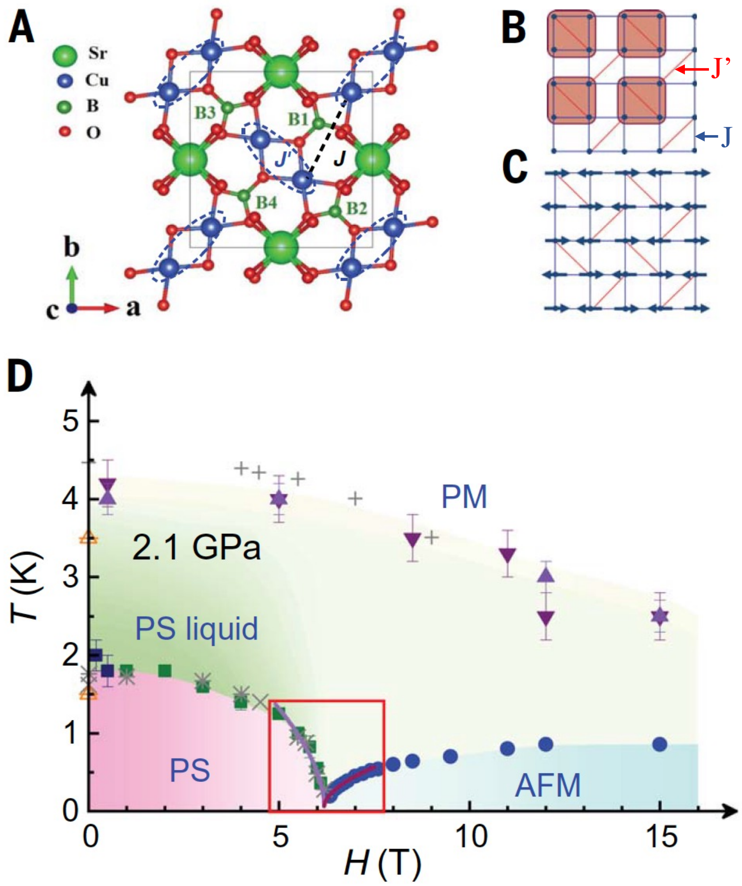

Figure 1: A - Top view of the layers of $SrCu_2(BO_3)_2$; B - Sketch of the full plaquette phase on the Shastry-Sutherland lattice; C - Sketch of Néel order on the Shastry-Sutherland lattice; D - Phase diagram of $SrCu_2(BO_3)_2$ as a function of field and temperature, as deduced from NMR (solid symbols) and specific heat measurements (other symbols). For details, see Ref.[11]. The pressure of 2.1 GPa ensures that the system is in the intermediate plaquette phase in zero field. The Ising critical temperature of the plaquette phase and the Néel temperature of the antiferromagnetic phase meet at a very low temperature, suggesting the proximity of a continuous transition that could be a deconfined quantum critical point. After Ref.[11].

In view of the difficulties faced by analytical or numerical approaches in reaching definitive conclusions, it would be most helpful to find an experimental system in which this question can be addressed. Quantum magnets in which a parameter can be tuned to induce a phase transition between phases with different symmetries are not so common. The natural control parameter to change magnetic couplings is pressure, but in transition metal oxides its effect is usually very small. One of the noticeable exceptions is the layered compound $SrCu_2(BO_3)_2$ (Fig. 1A), a system famous for its numerous magnetization plateaus[5]. This compound is an almost perfect realization of a 2D model known as the Shastry-Sutherland model[6], a model of orthogonal dimers (lattice of red dimer bonds with coupling J' and blue inter-dimer bonds with coupling J in Fig. 1B-C). For not too large inter-dimer coupling J, the ground state of this model is a product of singlet dimers (Fig. 2, left panel). Increasing the inter-

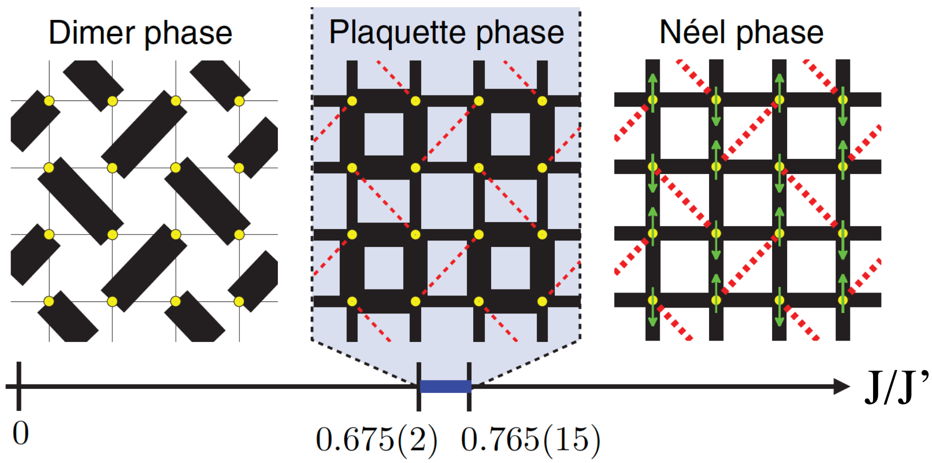

Figure 2: Zero-temperature phase diagram of the Shastry-Sutherland as a function of the ratio of the inter-dimer coupling to the intra-dimer coupling J/J'. After Ref.[7].

dimer coupling must ultimately lead to antiferromagnetic Néel order (Fig.2, right panel), but this transition takes place through an intermediate, twofold degenerate gapped phase known as the plaquette phase, in which strong bonds form around empty plaquettes of the Shastry-Sutherland model (Fig.2, middle panel). Luckily enough, the intra-dimer coupling in $SrCu_2(BO_3)_2$ corresponds to a Cu-O-Cu path close to 90 degrees and turns out to be highly sensitive to pressure. In addition, at ambient pressure, the ratio of inter- to intra-dimer exchange couplings is equal to 0.63, close to boundary to the plaquette phase, and pressure has indeed been shown to induce a first-order transition into a twofold degenerate phase with a thermal Ising transition at 2K[8, 9]. In 2019, it has been suggested by Lee et al[10] that the transition into the Néel antiferromagnetic phase could be an example of a DQCP. However, the experimental situation is not so clear regarding the pressure needed to reach the AF phase, and even less clear regarding the nature of the phase transition. One major obstacle is that the pressure is not a parameter that one can easily tune continuously (experiments are typically done at a set of pressures), so to check something as subtle as the difference between a weakly first-order transition and a DQCP is a real challenge.

There is of course another well known control parameter in quantum magnets, the magnetic field. In a gapped phase, the gap Δ closes at a critical field given by $g\mu_B H_c = \Delta$, and, except in 1D, there is long-range magnetic order in the phase above H_c. A gapped phase does not need to break any symmetry, a well known example being the spin-1/2 ladder, and in that case the gap closing in a 2-dimensional system is a standard continuous transition in the 3D O(2) universality class. However, if the gapped phase breaks a lattice symmetry, as in $SrCu_2(BO_3)_2$, the transition induced by closing the gap with an external magnetic field could take place between two phases with unrelated symmetry breakings, hence be a candidate for a DQCP. This is precisely the point of view adopted by Cui et al[11], and the measurements they have reported at 2.1 GPa (see Fig. 1D) as a function of magnetic field suggest that a transition indeed takes place between two phases with unrelated symmetries:

Upon increasing the field, the Ising critical temperature decreases to reach a minimum at extremely low temperature. Beyond that field, another critical temperature develops that they attribute to magnetic long-range order based on NMR evidence.

Unfortunately, the transition between these phases appears to be first order at very low temperature: The two critical lines meet at a small but strictly positive temperature that is also the end point of a first-order transition line that starts at zero temperature. This is not a final blow though. By repeating the experiments at 2.4 GPa, they found a similar phase diagram but with a reduced temperature at which the critical lines meet. If this temperature was to go to zero by further increasing the pressure, the resulting transition would be a continuous quantum transition between phases that break unrelated symmetries, hence a good candidate for a DQCP. This led the authors to speculate that there is indeed a DQCP in the proximity of the experimental range they could reach so far. As a first step towards testing this conjecture, they have analyzed scaling relations in the vicinity of this first order transition, and they appear to be consistent with the expectations for a DQCP.

Clearly, the next step will be to perform experiments at larger pressure to see if one can turn this transition into a continuous one. This is quite challenging because, on top of the difficulty of working at such high pressures, there is the problem of guessing the right pressure - if there is a multicritical DQCP, it will only be realized at one pressure, and since the pressure cannot be tuned continuously, it could take several experiments to hit a pressure close enough to the DQCP to observe a continuous phase transition. The situation might even be more complicated given the yet unclear nature of the intermediate phase of $SrCu_2(BO_3)_2$ (see below). In any case, this paper is to the best of my knowldege the first one to draw a clear road map towards the possible observation of a DQCP in a quantum magnet. It will be very interesting to see if further measurements will be able to confirm this prediction.

Now, even if the transition can be made continuous by increasing pressure, this will not be the end of the story. Indeed, if there is a DQCP, the next question will be to identify the fractional excitations that are confined in one phase and condense in the other one. For that, it is necessary to know the nature of the phases on both sides of the transition. It turns out that this is less simple than one might hope. The first problem is the nature of the intermediate phase. The Shastry-Sutherland model has proven to be able to account for the entire phase diagram of $SrCu_2(BO_3)_2$ as a function of temperature, pressure and field *except* for one phase, the intermediate phase. Indeed, in the plaquette phase of the Shastry-Sutherland model, all Cu sites remain equivalent, but in the intermediate phase of $SrCu_2(BO_3)_2$, there are two Cu sites, as revealed early on by NMR[12]. There is some experimental evidence that this phase is probably the full-plaquette phase[13], where singlets form on the plaquettes that have a diagonal coupling rather than on the empty plaquettes (see Fig. 1B). This terminology is however a bit misleading. In fact, this phase is very different from the plaquette phase of the Shastry-Sutherland model because it induces a difference between the dimers pointing in one direction and those pointing perpendicular to that direction, resulting in two different gaps. If anything, this phase should rather be thought of a set of spin-1 chains, half the dimers being in a triplet state while the other half remain in a singlet state[14]. In these conditions, the nature of the fractional excitations that would deconfine at the transition must still be worked out.

The nature of the antiferromagnetic phase is not obvious either. The gap closing could very well correspond to closing the gap of the spin-1 chains while leaving the gap of the other dimers open. The NMR spectrum would then be the superposition of a split spectrum for the magnetic sites, and a broader spectrum for the non magnetic sites. The NMR data reported in Fig. 4 of Ref. [11] show a clear splitting, consistent with antiferromagnetic order, but the split line seems to sit on top of a broad background that could originate from non magnetic sites. This is a crucial issue because, if the symmetry that is broken in the intermediate phase remains broken in the antiferromagnetic phase, the phase transition would be a standard field induced gap closing. However, the fact that the Ising transition temperature drops upon approaching the field where the gap closes suggests that the Ising symmetry that is broken in the intermediate phase is no longer broken in the antiferromagnetic phase, supporting the proposal by Cui et al[11] that the transition could be a DQCP if it becomes continuous.

All these questions point to the necessity of carrying on further experiments at larger pressure to see among other things if the phase that is reached at intermediate pressure when closing the gap is indeed the Néel phase expected at very high pressure. It would also be very useful if theorists could identify the right model that describes all the properties of $SrCu_2(BO_3)_2$, including those of the intermediate phase. This might require to include phonons since this intermediate phase is not that of the purely magnetic model with inter- and intra-dimer exchange.

References

[1] T. Senthil, A. Vishwanath, L. Balents, S. Sachdev, M. P. A. Fisher, Science 303, 1490–1494 (2004).

[2] A. W. Sandvik, Phys. Rev. Lett. 98, 227202 (2007).

[3] A. Nahum, J. T. Chalker, P. Serna, M. Ortuño, A. M. Somoza, Phys. Rev. X 5, 041048 (2015).

[4] B. Zhao, J. Takahashi, A. W. Sandvik, Phys. Rev. Lett. 125, 257204 (2020).

[5] For an early review, see S. Miyahara and K. Ueda, J. Phys.: Condens. Matter 15, R327 (2003).

[6] B. S. Shastry and B. Sutherland, Physica B 108, 1069 (1981).

[7] P. Corboz and F. Mila, Phys. Rev. B 87, 115144 (2013).

[8] J. Guo, G. Sun, B. Zhao, L. Wang, W. Hong, V. A. Sidorov, Nv. Ma, Q. Wu, S. Li, Z. Y. Meng, A. W. Sandvik, and L. Sun, Phys. Rev. Lett. 124, 206602 (2020).

[9] J. Larrea Jiménez, S. P. G. Crone, E. Fogh, M. E. Zayed, R. Lortz, E. Pomjakushina, K. Conder, A. M. Läuchli, L. Weber, S. Wessel, A. Honecker, B. Normand, Ch. Rüegg,, P. Corboz, H. M. Rønnow ,and F. Mila, Nature 592, 370 (2021).

[10] J. Y. Lee, Y.-Z. You, S. Sachdev, A. Vishwanath, Phys. Rev. X 9, 041037 (2019).

[11] Y. Cui, L. Liu, H. Lin, K.-H. Wu, W. Hong, X. Liu, C. Li, Z. Hu, N. Xi, S. Li, R. Yu, A. W. Sandvik, W.dYu, Science 380, 1179 (2023).

[12] T. Waki, K. Arai, M. Takigawa, Y. Saiga, Y. Uwatoko, H. Kageyama, and Y. Ueda, J. Phys. Soc. Japan 76, 073710 (2007).

[13] M. E. Zayed, C. Rüegg, J. Larrea J., A. M. Läuchli, C. Panagopoulos, S. S. Saxena, M. Ellerby, D. F. McMorrow, T. Strässle, S. Klotz, G. Hamel, R. A. Sadykov, V. Pomjakushin, M. Boehm, M. Jiménez-Ruiz, A. Schneidewind, E. Pomjakushina, M. Stingaciu, K. Conder, and H. M. Rønnow, Nat. Phys. 13, 962 (2017).

[14] M. Moliner, I. Rousochatzakis, and F. Mila, Phys. Rev. B 83, 140414(R) (2011).