Rapid Communications

Competition between intermediate plaquette phases in $SrCu_2(BO_3)_2$ under pressure

C. Boos,[1,2,*] S. P. G. Crone,[3] I. A. Niesen,[3] P. Corboz,[3] K. P. Schmidt,[1,†] and F. Mila[2,‡]

[1]*Institute for Theoretical Physics, FAU Erlangen-Nürnberg, Germany*

[2]*Institute of Physics, Ecole Polytechnique Fédérale de Lausanne (EPFL), CH 1015 Lausanne, Switzerland*

[3]*Institute for Theoretical Physics and Delta Institute for Theoretical Physics, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands*

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Building on the growing evidence based on NMR, magnetization, neutron scattering, electron spin resonance, and specific heat that, under pressure, $SrCu_2(BO_3)_2$ has an intermediate phase between the dimer and the Néel phase, we study the competition between two candidate phases in the context of a minimal model that includes two types of intra- and interdimer interactions without enlarging the unit cell. We show that the empty plaquette phase of the Shastry-Sutherland model is quickly replaced by a quasi-one-dimensional full plaquette phase when intra- and/or interdimer couplings take different values, and that this full plaquette phase is in much better agreement with available experimental data than the empty plaquette one.

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Almost two decades after the discovery of the first magnetization plateaus, the investigation of the layered material $SrCu_2(BO_3)_2$ under extreme conditions continues to attract a lot of attention and to reveal new fascinating properties. If there is by now ample evidence in favor of a sequence of magnetization plateaus at 1/8, 2/15, 1/6, 1/4, 1/3, and 1/2 (and possibly 2/5) [1–10], the structure of some of these plateaus remains debated, and several groups are attempting to perform x-ray or inelastic neutron scattering (INS) in fields above 27 Tesla and at very low temperature to have direct information on the structure of the 1/8 plateau. In parallel, the investigation of the phase diagram under pressure using various techniques ranging from NMR [11] to magnetization [10], electron spin resonance (ESR) [12], INS [13], and specific heat [14] has revealed the presence of a phase transition at around 1.7 GPa to a new gapped phase that is the subject of the present Rapid Communication.

$SrCu_2(BO_3)_2$ is described to a very good accuracy by the stacking of the two-dimensional (2D) Shastry-Sutherland model [15], also known as the orthogonal dimer model [16], defined by the Hamiltonian

$$H = J \sum_{\langle\langle i,j \rangle\rangle} \vec{S}_i \cdot \vec{S}_j + J' \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j, \qquad (1)$$

where J is the intradimer coupling and J' the interdimer coupling. In the limit $J' = 0$, the system consists of a set of decoupled dimers, and the exact ground state is a product of singlets on these dimers. Due to the frustrated nature of the interdimer coupling, this remains strictly true as long as J' is not too large. In the opposite limit $J = 0$, the system is a square lattice with nearest-neighbor antiferromagnetic couplings, and the ground state possesses long-range Néel order. In between, there is an intermediate phase that, after some debate [17–25], has been convincingly proven to be the empty plaquette phase (EPP) depicted in Fig. 1(b) and to exist in the range $0.675(2) < J'/J < 0.765(15)$ [26]. The dominant interlayer interactions are not expected to change the physics qualitatively since the product of dimer singlets is still a ground state, while the small Dzyaloshinskii-Moriya interactions are not expected to shift the boundaries significantly since their effect on the ground-state energy of a gapped singlet phase is of second order.

Since up to an overall energy scale the ratio J'/J is the only parameter of the Shastry-Sutherland model, applying hydrostatic pressure to change this ratio is a natural way to probe this phase diagram, and this has been first attempted in 2007 using NMR [11]. This experiment has indeed revealed the presence of a new phase at 2.4 GPa, but in this intermediate phase, there are two types of Cu sites. This is incompatible with the EPP, in which all Cu sites remain equivalent. The report of a weak orthorhombic distortion already at low pressure has led to the investigation of a model with two sets of intradimer couplings [27]. If the couplings are sufficiently different, another intermediate phase is realized. It is a one-dimensional phase related to a spin-1 Haldane chain. Note, however, that the presence of an orthorhombic distortion at low pressure has not been confirmed by subsequent experiments.

More recently, neutron scattering experiments have confirmed the presence of an intermediate phase [13] characterized by the presence of an additional second excitation branch at low energies, in sharp contrast with the dimer phase. The structure factors of these excitations appear to be incompatible with the EPP, another indication that the intermediate phase is not that of the Shastry-Sutherland model. They are, however, compatible with a putative full plaquette phase (FPP) in which bonds get stronger around plaquettes with diagonal couplings [see Fig. 1(c)].

In this Rapid Communication, we discuss theoretically the possible nature of this intermediate phase. We show that the

*carolin.boos@fau.de

†kai.phillip.schmidt@fau.de

‡frederic.mila@epfl.ch

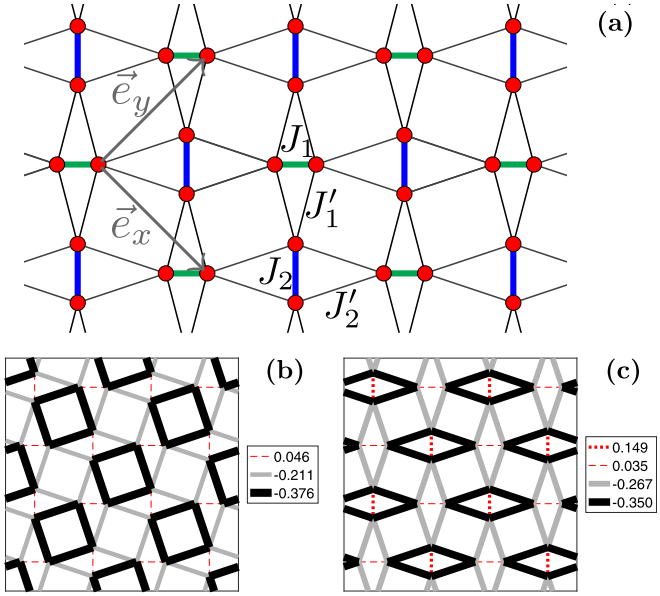

FIG. 1. (a) Sketch of the distorted orthogonal dimer lattice. The unit cell is defined by the unit vectors $\vec{e}_x$ and $\vec{e}_y$. (b) Spin-spin correlations in the EPP and (c) in the FPP phase obtained with iPEPS for the parameters used in Figs. 3(a) and 3(b) and 3(d) and 3(e), respectively.

Haldane and the FPP actually build a single phase in the phase diagram of a generalized Shastry-Sutherland model that includes two types of intra- and interdimer couplings, and that the properties of this phase are in much better agreement with available experimental data than those of the EPP. Consequences for the three-dimensional system are briefly discussed.

In choosing the model to describe the competition between different possible intermediate phases, we have paid special attention to the fact that, so far, no distortion could be detected (although there has to be one of course, as discussed at the end of the Rapid Communication). So we have concentrated on a minimal modification that contains two sets of inequivalent J bonds, J_1 and J_2, as assumed in Ref. [27], but also two sets of inequivalent J' bonds, J_1' and J_2' [see Fig. 1(a)]. Models with inequivalent J' bonds have been introduced in Ref. [22] as starting points of series expansions (SEs), but the relative stability of the EPP and FPP phases has not been studied. The first goal of the present Rapid Communication is to map out precisely these stability regions. As pointed out recently by Lee *et al.* [28], the two candidate plaquette phases correspond to natural distortions in a Landau expansion, depending on the sign of the coupling constant. In the FPP, diamonds with short intra- and interdimer bonds form. Naively one could expect both intra- and interdimer couplings to get stronger, but this is not the case. The intradimer coupling corresponds to a Cu-O-Cu bond with an angle of 97.6°, and making it shorter will actually *decrease* the magnitude of the coupling constant [12]. By contrast, the interdimer coupling is a more standard geometry, and the coupling constant is expected to get stronger if the bond gets shorter. So we have considered the parameter range where the weaker intradimer coupling J_1 is surrounded by stronger interdimer couplings J_1'. Taking the other configuration would anyway require very different interdimer bonds to stabilize the FPP, a possibility which is not realistic.

Our results have been obtained by two complementary methods, infinite projected entangled-pair states (iPEPS) and high-order SEs. An iPEPS is a variational tensor-network ansatz for two-dimensional ground states in the thermodynamic limit [29–31], where the accuracy is systematically controlled by the bond dimension D of the tensors. This approach has already been successfully applied in previous studies of the Shastry-Sutherland model (see, e.g., Refs. [26,32]). The SE for the ground-state energies of the EPP and FPP were performed by the Löwdin algorithm [33–35] while the energies of the elementary triplon excitations [36,37] and the dynamic structure factors have been determined using perturbative continuous unitary transformations [38,39]. In all cases we introduce a deformation parameter λ so that the unperturbed part $\lambda = 0$ corresponds to isolated (empty or filled) plaquettes and $\lambda = 1$ to the distorted Shastry-Sutherland model under study. The ground-state energy for the EPP (FPP) is calculated up to order 9 (8) in λ. The excitation energies of single triplons have been determined up to order 6 in both plaquette phases. The static and dynamic structure factors are calculated up to order 5. The derived orders are similar to other plaquette expansions [22,25,40,41]. For the distorted Shastry-Sutherland model we increased the maximal perturbative order of the ground-state energies by two compared to Ref. [22]. To our knowledge, the dynamic structure factor was not calculated before. All series are extrapolated up to $\lambda = 1$ using Padé extrapolation [42]. In the following we use the variance of the different Padé extrapolants as uncertainty of the extrapolation. For details about both methods, see the Supplemental Material [37] (see also Refs. [43–64]).

Since we have four parameters, hence three up to an overall energy scale, plotting the full phase diagram is tricky. We have chosen to study the phase diagram in three planes defined by $J_1' = J_2' = J'$ [Fig. 2(a)], $J_1 = J_2 = J$ [Fig. 2(b)], and $J_2'/J_2 = 0.68$ [Fig. 2(c)]. In the phase diagram of Fig. 2(a), we revisit the effect of different intradimer couplings discussed in Ref. [27]. Qualitatively, the results are the same, with four phases (dimer, EPP, Néel, and Haldane), but the extent of the EPP is considerably reduced, and accordingly the Haldane phase is stabilized in a much larger parameter range that extends up to $J_1/J_2 \simeq 0.98$, very close to the isotropic point. In the phase diagram of Fig. 2(b), we study the effect of different interdimer couplings. This phase diagram shows that the EPP is indeed the only one appearing in the isotropic Shastry-Sutherland model, but that it only takes a modest difference to stabilize the FPP. Finally, in Fig. 2(c), we show a cut in which both the ratios J_1/J_2 and J_2'/J_1' vary for a fixed value of $J_2'/J_2 = 0.68$. At the bottom left corner, the Haldane phase has to be stabilized, as we know from Fig. 2(a), while at the top right corner, it is the FPP that is stabilized, as is clear from Fig. 2(b). Quite remarkably, there is no phase transition between them, and these two phases actually constitute a single quasi-one-dimensional phase in which strong correlations are concentrated around full plaquettes [see Fig. 1(c)]. Similar correlations have actually already been reported in the bottom right panel of Fig. 3 of Ref. [27].

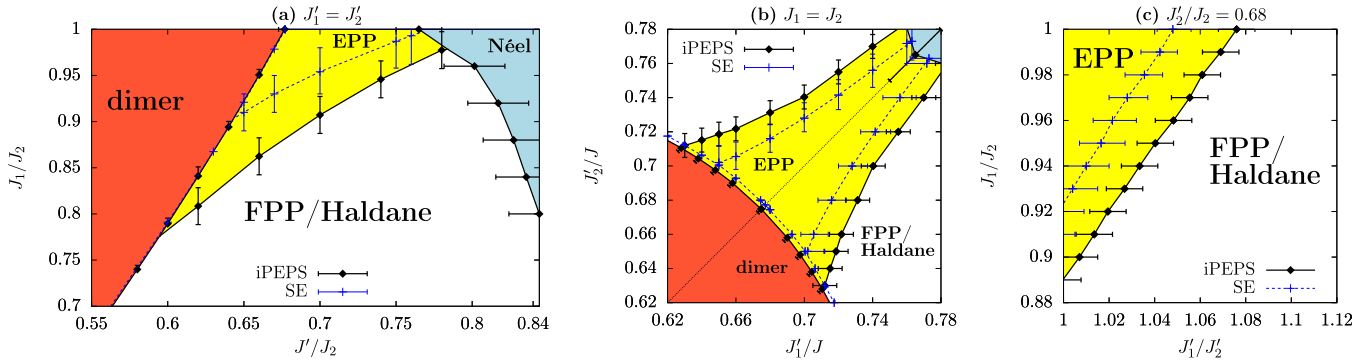

FIG. 2. Ground-state phase diagrams of distorted Shastry-Sutherland models by iPEPS and SE. In (a) for identical nearest-neighbor couplings $J_1' = J_2'$, in (b) for identical diagonal couplings, and in (c) both asymmetries are included at the ratio $J_2'/J_2 = 0.68$. The area based on the iPEPS results of the dimer singlet phase is colored in red, the EPP in yellow, the FPP/Haldane phase in white, and the Néel phase is shown in blue.

To further demonstrate that the FPP and the Haldane phase are adiabatically connected we have computed the inter- and intradimer spin-spin correlations and the correlation lengths along a linear path in parameter space connecting the model with unequal interdimer couplings ($J_2'/J_2 = 0.66$, $J_1'/J_2' = 1.1$, $J_1/J_2 = 1$) to the one with unequal intradimer couplings ($J_2'/J_2 = 0.55$, $J_1'/J_2' = 1$, $J_1/J_2 = 0.5$). The iPEPS results ($D = 10$ full update simulations) given in the Supplemental Material [37] show that all correlations change smoothly, i.e., that there is no sign of a quantum phase transition along this path. Interestingly, the ratio of the correlation lengths in the x and y direction, ξ_x/ξ_y, remains almost constant along this path, revealing the anisotropic nature of this phase, that we will now call the FPP/Haldane phase, even in the limit of equal intradimer couplings ($J_1 = J_2$).

Let us turn to the properties of the EPP and the FPP/Haldane phase. As mentioned above, the first indication that the EPP phase cannot be the intermediate phase came from NMR [11], which detected two types of Cu sites. Since NMR is (by necessity) performed in a finite magnetic field, it is interesting to look for complementary evidence in zero-field experiments, ESR, neutron scattering, and specific heat [12–14]. All these experiments confirm the presence of two well-defined magnetic excitations; one at an energy comparable to that of the gap in the dimer phase just before the transition (ESR, neutron scattering), and one at an energy about two times smaller (neutron scattering, specific heat). The neutron-scattering measurements have followed the dispersion along the line ($k_x, k_y = 0$) in the Brillouin zone, while the specific heat could keep track of the pressure dependence of the gap, i.e., of the minimum of the lowest excitation, with clear evidence that it decreases with pressure. Neutron scattering also revealed that the structure factors of the two low-lying excitations have different momentum dependencies.

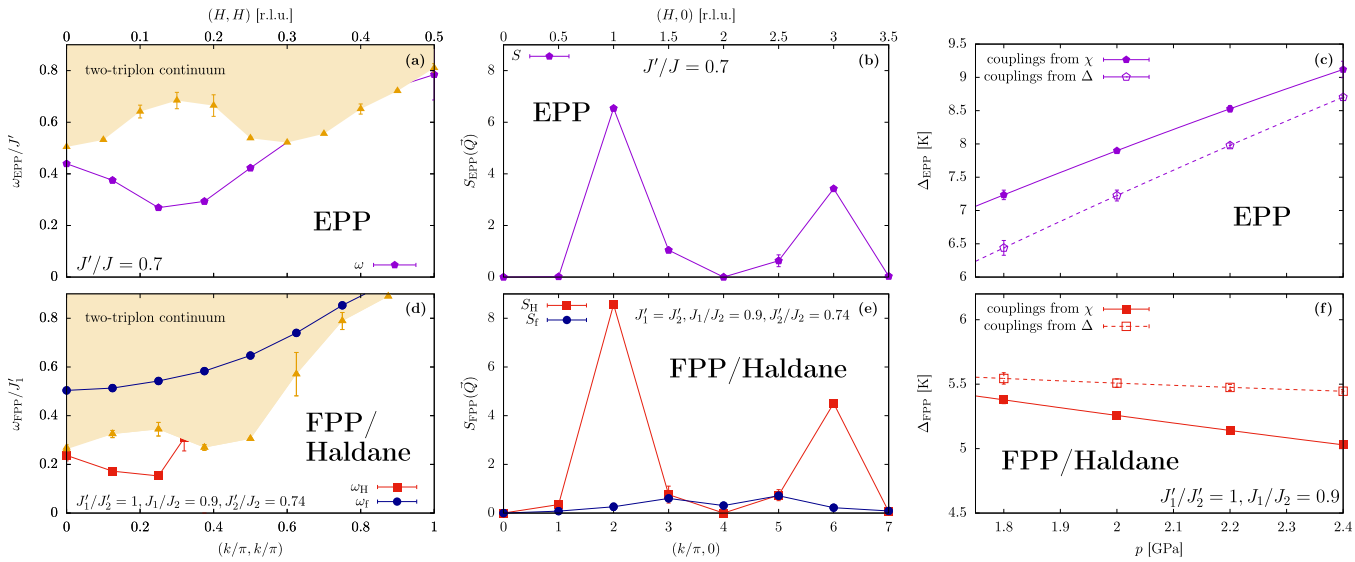

FIG. 3. Magnetic excitations in the EPP (top) and FPP/Haldane (bottom). Panels (a) and (d): magnetic excitations along $k_x = k_y$; panels (b) and (e): static structure factors along $k_y = 0$. The parameters are given inside the figures. Panels (c) and (f): pressure dependence of the gap (with couplings from magnetic susceptibility χ [13] and excitation energies Δ measured by ESR [12]; see main text). All lines are guides to the eye.

To make contact with these experiments, we have studied the magnetic excitations in both phases. Characteristic results are plotted in the top panels [Figs. 3(a)–3(c)] for the EPP and in the bottom panels [Figs. 3(d)–3(f)] for the FPP. In the EPP there is a single low-energy excitation that can be interpreted as the dispersion of a triplet plaquette in a sea of singlet plaquettes. For small to intermediate ratios J'/J its dispersion has a minimum along the direction $k_x = k_y$. Since ESR only measures the zero-momentum excitations while the specific heat detects the gap, i.e., the minimal energy, this dispersion could be compatible with ESR and specific heat. However, this is not possible if the ratio between both energies is slightly larger than 2, since then the mode at $\vec{k} = (0, 0)$ decays. The structure factor matches that of the lowest excitation detected in neutron scattering. However, this possibility is excluded by neutron scattering, which has observed two well-defined excitations at the same momentum. In addition, the energy gap in the EPP increases with pressure as shown in Fig. 3(f), where the pressure is introduced by changing the ratio J'/J in the isotropic model following [12,13]. This is in clear contradiction with specific heat data. Note that this remains true also for the EPP in a model with stronger couplings around one set of empty plaquettes, which corresponds to the intrinsic lattice distortion of that phase [28].

In the FPP/Haldane phase, the situation is very different. There are two well-defined excitations. The dispersions ω_{H} and ω_{f} along $k_x = k_y$, which are protected by local symmetries, are shown in Fig. 3(d), and, at least not too far from the Néel phase, the lowest one has an energy about half that of the other one at $k = 0$. For momenta $k_x = k_y \leqslant 0.5\pi$ we determine accurately the lower bound of the two-triplon continuum, whereas for larger momenta the true continuum might be below, so the excitation ω_{H} decays. Details about the determination of the continuum are given in the Supplemental Material [37]. The structure factors of these excitations along $k_y = 0$ are shown in Fig. 3(e), where we can exclude decay for momenta $k_x/\pi = \{0, 2, 4, 6\}$. They are in good agreement with neutron scattering, which resolved a lower excitation with a large intensity at $k_x/\pi = 2$ and a very small intensity at $k_x/\pi = 4$, and an excitation at larger energy with a rather small intensity for all momenta [see Fig. 3(f) of Ref. [13]]. In addition, the gap decreases with pressure, which matches with specific heat data. This conclusion has been reached following a path in parameter space assuming $J_1 = 0.9J_2$ and adjusting the average of J_1 and J_2 to the estimates from INS for a symmetric model, but we have checked that the sign of the slope remains negative for similar paths. So the case in favor of the FPP appears to be very strong.

The presence of two low-lying excitations in the FPP/Haldane phase can be traced back to the quasi-one-dimensional nature of this phase. In the limit of completely decoupled chains ($J_2' = 0$), the branch called Haldane corresponds to the triplet excitation branch of a spin-1 chain, realized when all the weak J_1 dimers are in a triplet state, while the branch called flat, which is indeed completely flat in that limit, corresponds to a singlet dimer on one of the weak J_1 bonds [37].

Next we briefly discuss the implications of the present results for the intermediate phase of $SrCu_2(BO_3)_2$. Within the minimal model studied in this Rapid Communication (a purely 2D model with two types of intra- and interdimer couplings), there is a single alternative to the EPP of the Shastry-Sutherland model, namely, a quasi-1D phase with strong correlations around full plaquettes, and the properties of this phase appear to be consistent with available data. If the system was purely 2D, the stabilization of this phase would induce an orthorhombic distortion since the C_4 symmetry is lost. This can be expected to remain true for $SrCu_2(BO_3)_2$, which is a three-dimensional crystal, if, in all layers, the weak intradimer couplings are oriented in the same direction. However, if this direction alternates from one layer to the next, the distortion is not expected any more to be a clear orthorhombic distortion, but to be some local rearrangement inside an essentially unchanged unit cell. The failure so far to detect any clear lattice distortion in the intermediate phase points to the second possibility with alternating directions.

There is also an interesting conceptual difference between the two plaquette phases regarding the nature of the phase transition. The EPP is an instability of the Shastry-Sutherland model that spontaneously breaks the symmetry even if all intra- and interdimer couplings remain the same. By contrast, the FPP is not an instability of the Shastry-Sutherland model. Like a spin-Peierls transition in spin-1/2 chains, it has to be an instability of the coupled spin-lattice system. So, when applying pressure, if there is a direct transition between the dimer phase and the FPP, it has to take place below the critical ratio at which the transition to the EPP takes place in the Shastry-Sutherland model. Otherwise, there would first be a transition to the EPP. Current estimates of the ratio J'/J at 1.7 GPa from ESR and susceptibility are in the range 0.66–0.665 [12–14], indeed below the critical ratio 0.675 of the EPP.

What could be the next step to confirm (or discard) the FPP as the intermediate phase of $SrCu_2(BO_3)_2$? Of course, a direct identification of the structural distortion would be ideal, but even if the distortion turns out to be too small to be detected, one could hope to detect it indirectly through selection rules. In that respect, measuring the phonons with Raman scattering as a function of pressure could be very helpful. Alternatively, since in our calculations the details of the excitation spectrum change significantly inside the intermediate phases, additional inelastic neutron-scattering measurements would be most welcome. Finally, a theoretical investigation of the properties of the intermediate phase in a magnetic field to make contact with NMR and with magnetization measurements is clearly needed. Work is in progress along these lines.

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[1] H. Kageyama, K. Yoshimura, R. Stern, N. V. Mushnikov, K. Onizuka, M. Kato, K. Kosuge, C. P. Slichter, T. Goto, and Y. Ueda, Phys. Rev. Lett. **82**, 3168 (1999).

[2] K. Onizuka, H. Kageyama, Y. Narumi, K. Kindo, Y. Ueda, and T. Goto, J. Phys. Soc. Jpn. **69**, 1016 (2000).

[3] K. Kodama, M. Takigawa, M. Horvatić, C. Berthier, H. Kageyama, Y. Ueda, S. Miyahara, F. Becca, and F. Mila, Science **298**, 395 (2002).

[4] M. Takigawa, K. Kodama, M. Horvatić, C. Berthier, H. Kageyama, Y. Ueda, S. Miyahara, F. Becca, and F. Mila, Phys. B (Amsterdam, Neth.) **346-347**, 27 (2004).

[5] F. Levy, I. Sheikin, C. Berthier, M. Horvatić, M. Takigawa, H. Kageyama, T. Waki, and Y. Ueda, Europhys. Lett. **81**, 67004 (2008).

[6] S. E. Sebastian, N. Harrison, P. Sengupta, C. D. Batista, S. Francoual, E. Palm, T. Murphy, N. Marcano, H. A. Dabkowska, and B. D. Gaulin, Proc. Natl. Acad. Sci. U.S.A. **105**, 20157 (2008).

[7] M. Jaime, R. Daou, S. A. Crooker, F. Weickert, A. Uchida, A. E. Feiguin, C. D. Batista, H. A. Dabkowska, and B. D. Gaulin, Proc. Natl. Acad. Sci. USA. **109**, 12404 (2012).

[8] M. Takigawa, M. Horvatić, T. Waki, S. Krämer, C. Berthier, F. Lévy-Bertrand, I. Sheikin, H. Kageyama, Y. Ueda, and F. Mila, Phys. Rev. Lett. **110**, 067210 (2013).

[9] Y. H. Matsuda, N. Abe, S. Takeyama, H. Kageyama, P. Corboz, A. Honecker, S. R. Manmana, G. R. Foltin, K. P. Schmidt, and F. Mila, Phys. Rev. Lett. **111**, 137204 (2013).

[10] S. Haravifard, D. Graf, A. E. Feiguin, C. D. Batista, J. C. Lang, D. M. Silevitch, G. Srajer, B. D. Gaulin, H. A. Dabkowska, and T. F. Rosenbaum, Nat. Commun. **7**, 11956 (2016).

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

[12] T. Sakurai, Y. Hirao, K. Hijii, S. Okubo, H. Ohta, Y. Uwatoko, K. Kudo, and Y. Koike, J. Phys. Soc. Jpn. **87**, 033701 (2018).

[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] J. Guo, G. Sun, B. Zhao, L. Wang, W. Hong, V. A. Sidorov, N. Ma, Q. Wu, S. Li, Z. Y. Meng, A. W. Sandvik, and L. Sun, arXiv:1904.09927.

[15] B. S. Shastry and B. Sutherland, Physica B+C **108**, 1069 (1981).

[16] S. Miyahara and K. Ueda, J. Phys.: Condens. Matter **15**, R327 (2003).

[17] M. Albrecht and F. Mila, Europhys. Lett. **34**, 145 (1996).

[18] S. Miyahara and K. Ueda, Phys. Rev. Lett. **82**, 3701 (1999).

[19] Zheng Weihong, C. J. Hamer, and J. Oitmaa, Phys. Rev. B **60**, 6608 (1999).

[20] E. Müller-Hartmann, R. R. P. Singh, C. Knetter, and G. S. Uhrig, Phys. Rev. Lett. **84**, 1808 (2000).

[21] A. Koga and N. Kawakami, Phys. Rev. Lett. **84**, 4461 (2000).

[22] Y. Takushima, A. Koga, and N. Kawakami, J. Phys. Soc. Jpn. **70**, 1369 (2001).

[23] C. H. Chung, J. B. Marston, and S. Sachdev, Phys. Rev. B **64**, 134407 (2001).

[24] A. Läuchli, S. Wessel, and M. Sigrist, Phys. Rev. B **66**, 014401 (2002).

[25] W. Zheng, J. Oitmaa, and C. J. Hamer, Phys. Rev. B **65**, 014408 (2001).

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

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

[28] J. Y. Lee, Y.-Z. You, S. Sachdev, and A. Vishwanath, arXiv:1904.07266.

[29] F. Verstraete and J. I. Cirac, arXiv:cond-mat/0407066.

[30] Y. Nishio, N. Maeshima, A. Gendiar, and T. Nishino, arXiv:cond-mat/0401115.

[31] J. Jordan, R. Orús, G. Vidal, F. Verstraete, and J. I. Cirac, Phys. Rev. Lett. **101**, 250602 (2008).

[32] P. Corboz and F. Mila, Phys. Rev. Lett. **112**, 147203 (2014).

[33] P. Löwdin, J. Chem. Phys. **19**, 1396 (1951).

[34] P. Löwdin, J. Math. Phys. **3**, 969 (1962).

[35] T. Kato, Phys. Rev. **77**, 413 (1950).

[36] K. P. Schmidt and G. S. Uhrig, Phys. Rev. Lett. **90**, 227204 (2003).

[37] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.100.140413 for details on the iPEPS, the SE, the excitations of the two-dimensional distorted Shastry-Sutherland model, and the orthogonal dimer chain.

[38] C. Knetter and G. S. Uhrig, Eur. Phys. J. B **13**, 209 (2000).

[39] C. Knetter, K. P. Schmidt, and G. S. Uhrig, J. Phys. A: Math. Gen. **36**, 7889 (2003).

[40] M. Arlego and W. Brenig, Phys. Rev. B **78**, 224415 (2008).

[41] M. Arlego and W. Brenig, Phys. Rev. B **84**, 134426 (2011).

[42] A. C. Guttmann, *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. Lebowitz (Academic, New York, 1989), Vol. 13.

[43] S. Singh, R. N. C. Pfeifer, and G. Vidal, Phys. Rev. B **83**, 115125 (2011).

[44] B. Bauer, P. Corboz, R. Orús, and M. Troyer, Phys. Rev. B **83**, 125106 (2011).

[45] H. C. Jiang, Z. Y. Weng, and T. Xiang, Phys. Rev. Lett. **101**, 090603 (2008).

[46] P. Corboz, R. Orús, B. Bauer, and G. Vidal, Phys. Rev. B **81**, 165104 (2010).

[47] H. N. Phien, J. A. Bengua, H. D. Tuan, P. Corboz, and R. Orús, Phys. Rev. B **92**, 035142 (2015).

[48] P. Corboz, Phys. Rev. B **94**, 035133 (2016).

[49] T. Nishino and K. Okunishi, J. Phys. Soc. Jpn. **65**, 891 (1996).

[50] R. Orús and G. Vidal, Phys. Rev. B **80**, 094403 (2009).

[51] P. Corboz, S. R. White, G. Vidal, and M. Troyer, Phys. Rev. B **84**, 041108(R) (2011).

[52] P. Corboz, T. M. Rice, and M. Troyer, Phys. Rev. Lett. **113**, 046402 (2014).

[53] P. Corboz, Phys. Rev. B **93**, 045116 (2016).

[54] T. Nishino, K. Okunishi, and M. Kikuchi, Phys. Lett. A **213**, 69 (1996).

[55] H.-Y. Yang, A. M. Läuchli, F. Mila, and K. P. Schmidt, Phys. Rev. Lett. **105**, 267204 (2010).

[56] J. Röchner, L. Balents, and K. P. Schmidt, Phys. Rev. B **94**, 201111(R) (2016).

[57] K. Coester and K. P. Schmidt, Phys. Rev. E **92**, 022118 (2015).

[58] R. W. Smith and D. A. Keszler, J. Solid State Chem. **93**, 430 (1991).
[59] J. Richter, N. B. Ivanov, and J. Schulenburg, J. Phys.: Cond. Matter **10**, 3635 (1998).
[60] A. Koga, K. Okunishi, and N. Kawakami, Phys. Rev. B **62**, 5558 (2000).
[61] N. B. Ivanov and J. Richter, Phys. Lett. A **232**, 308 (1997).
[62] A. Honecker, S. Wessel, R. Kerkdyk, T. Pruschke, F. Mila, and B. Normand, Phys. Rev. B **93**, 054408 (2016).
[63] S. R. White and D. A. Huse, Phys. Rev. B **48**, 3844 (1993).
[64] O. Golinelli, Th. Jolicoeur, and R. Lacaze, Phys. Rev. B **50**, 3037 (1994).