Rise and fall of plaquette order in Shastry-Sutherland magnet revealed by pseudo-fermion functional renormalization group

Ahmet Keles\textsuperscript{1,}\textsuperscript{*} and Erhai Zhao\textsuperscript{2}

\textsuperscript{1}Department of Physics, George Mason University, Fairfax, Virginia 22030, USA
\textsuperscript{2}Department of Physics and Astronomy, Middle East Technical University, Ankara 06800, Turkey

The Shastry-Sutherland model as a canonical example of frustrated magnetism has been extensively studied. The conventional wisdom has been that the transition from the plaquette valence bond order to the Neel order is direct and potentially realizes a deconfined quantum critical point beyond the Ginzburg-Landau paradigm. This scenario was challenged recently by improved numerics from density matrix renormalization group which offers evidence for a narrow gapless spin liquid between the two phases. Prompted by this controversy and to shed light on this intricate parameter regime from a fresh perspective, we report high-resolution functional renormalization group analysis of the generalized Shastry-Sutherland model. The flows of over 50 million running couplings provide a detailed picture for the evolution of spin correlations as the frequency-energy scale is dialed from the ultraviolet to the infrared to yield the zero temperature phase diagram. The singlet dimer phase emerges as a fixed point, the Neel order is characterized by divergence in the vertex function, while the transition into and out of the plaquette order is accompanied by pronounced peaks in the plaquette susceptibility. The plaquette order is suppressed before the onset of the Neel order, lending evidence for a finite spin liquid region for $J_1/J_2 \in (0.77, 0.82)$, where the flow is continuous without any indication of divergence.

Forty years after the introduction of the Shastry-Sutherland (SS) model [1], its ground state phase diagram remains inconclusive. The model describes quantum spins on the square lattice with competing antiferromagnetic exchange interactions, $J_1$ for the horizontal/vertical bonds and $J_2$ for the decimated diagonal bonds connecting the empty plaquettes, see Fig. 1. Owing to the frustration, the model has long been suspected to host exotic ground states and phase transitions. A large body of theoretical works have established the existence of three phases, see e.g. [2] and [3, 4] for a synopsis of earlier and recent results, respectively. The $J_1 < J_2/2$ limit is exactly solvable and the ground state is a product state of diagonal dimers (spin singlets). For intermediate value of $J_1/J_2$, the ground state is a plaquette valence bond solid, while Neel order takes over for large $J_1/J_2$. The most interesting, and controversial, question regards the nature of the plaquette-to-Neel (pN) transition: is it conventional, a deconfined quantum critical point, or through an additional spin liquid phase?

Remarkably, the SS model has an almost ideal realization in SrCu$_2$(BO$_3$)$_2$ crystals, where phase transitions can be induced by tuning the hydrostatic pressure [5, 6]. Inelastic neutron scattering found signatures of the plaquette phase [7], and heat capacity measurements confirmed the dimer-to-plaquette transition [8, 9]. Yet a direct plaquette-to-Neel transition was not observed in the anticipated pressure range. These experiments renewed the effort to examine this intriguing region using the state-of-the-art numerical techniques. Earlier tensor network (tPEPS) calculations confirmed the plaquette phase within the region $J_1/J_2 \in [0.675, 0.765]$ [10–14] and a weak first order pN transition. A recent density matrix renormalization group (DMRG) study [3] with cylinders of circumference up to ten sites yielded similar phase boundary $J_1/J_2 \in [0.675, 0.77]$ but a continuous pN transition with spin correlations supporting a deconfined quantum critical point. Another DMRG using cylinder circumference up to 14 sites concludes that a spin liquid phase exists in the window $J_1/J_2 \in [0.79, 0.82]$ between the plaquette and the Neel phase [4]. A core difficulty in reaching a consensus is attributed to the near degeneracy of the competing orders in this region. The finite-size limitation of DMRG means that the ground state can only be inferred by extrapolation via careful finite size scaling analysis.

The size restriction prompts us to adopt an alternative approach diametrically opposed to exact diagonalization or DMRG on finite systems. The algorithm directly accesses the infrared and thermodynamic limit while treating all competing orders on equal footing without bias. It starts from the microscopic spin Hamiltonian and successively integrates out the higher frequency fluctuations with full spatial (or equivalently momentum) resolution retained at each step. The scale-dependent effective couplings and correlation functions are obtained by numerically solving the Functional Renormalization Group (FRG) flow equations [15–17]. As the frequency scale $\Lambda$ slides from $J_{1,2}$ down to zero, the zero temperature phase diagram is determined. Such FRG approach to quantum spin systems, first established in 2010 [18], has yielded insights for many frustrated spin models. But its application to the SS model has not been successful, perhaps due to two reasons. First, in contrast to the Neel order, the dimer or the plaquette order cannot be inferred naively from the divergence of vertex functions, making it challenging to locate their phase boundaries. Second, as we shall show below, the pN transition region is better understood by examining a generalized model that reduces to the SS model in a particular limit.

In this work, high resolution FRG analysis of the generalized SS model is achieved by overcoming these technical barriers. To maintain sufficient momentum and frequency resolution, one must keep track of millions of running couplings at each FRG step. The calculation is made possible by migrating to the GPU platform which led to performance improvement by orders of magnitude [19, 20]. Despite being a completely different approach, the phase boundaries predicted from our FRG are remarkably close to the state-of-the-art DMRG. The
agreement further establishes FRG as an accurate technique for frustrated quantum magnetism. Most importantly, the plaquette susceptibility from FRG indicates the plaquette order terminates around $J_1/J_2 \approx 0.77$ before the onset of weak Neel order around $J_1/J_2 \approx 0.82$. It supports the existence of a spin liquid region between the plaquette and Neel phase proposed in Ref. [4]. Thus, the SS model is a strong candidate to host spin liquid, and SrCu$_2$(BO$_3$)$_2$ offers exciting opportunity to realize and probe the elusive spin liquid phase.

**Model and pseudofermion FRG.**—Our starting point is the generalized Shastry-Sutherland Hamiltonian [21]

$$H = \kappa J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{i, \text{diag}} \mathbf{S}_i \cdot \mathbf{S}_j$$

where $\mathbf{S}_i$ are spin one-half operators ($S = 1/2$), $i, j$ label the sites, and $J_{1,2} > 0$ are antiferromagnetic exchange couplings. The first (second) sum is over nearest neighbors on the square lattice represented by the solid (dashed) black lines in Fig. 1(a), the last sum is over the alternating diagonal bonds indicated by the red lines. The original SS model corresponds to the limit $\kappa = 1$ [1, 2]. A small $\delta J_1 = (\kappa - 1) J_1$ acts as a source field to break the double degeneracy and favor valence bond order within the shaded plaquettes. It plays a crucial role in our analysis and facilitates the calculation of plaquette susceptibility. We will stay close to the limit $\kappa \to 1$ throughout.

To predict the phase diagram of Hamiltonian Eq. (1), FRG finds its generating functional, i.e., an effective field theory parametrized by self-energies, four-point and higher-order vertices, for each given frequency/energy scale $\Lambda$. The self-energies and vertices obey the formally exact flow equations that can be truncated and solved numerically. More specifically, the many-spin problem is first converted to an interacting fermion problem using the pseudofermion representation [18], $S^\mu = (1/2) \psi_i^\mu \psi_i$. Here $\sigma^\mu$ are the Pauli matrices, and $\psi_i$ annihilates a fermion at site $i$ with spin $\sigma = \uparrow, \downarrow$ etc.. The resultant fermion Hamiltonian only has quartic interactions but no kinetic energy term (the fermions are localized and constrained at one particle per site). So the bare single-particle Green function $G_0(\omega) = 1/\ii \omega$ with $\omega$ being the frequency [18]. Then the flow equations for the interacting fermion problem can be solved by generalizing the expansion and truncation schemes extensively benchmarked for strongly correlated electronic materials [22, 23].

The implementation of pseudofermion FRG are well documented in the original work [18] and later improvements [24–37]. A brief outline is as follows. Starting from an ultraviolet scale $\Lambda = \Lambda_{\text{UV}} \gg J_{1,2}$ and using the bare interaction in Eq. (1) and bare Green function to set up the initial condition, the coupled integro-differential equations for the scale-dependent self-energy $\Sigma^\omega(\omega)$ and four-point vertex $\Gamma^\omega_{\mu
u}(\omega_1', \omega_2')$ are solved successively in small steps along a discretized grid of the frequency/energy scale $\Lambda$ until it is reduced down to the infrared $\Lambda = \Lambda_{\text{IR}} \to 0$. During the flow, the self-energy $\Sigma^\Lambda(\omega)$ is renormalized to gain nontrivial frequency dependence as higher frequency fluctuations induce retarded interactions. But it remains site-independent, i.e. fermions hopping is prohibited. The four-point vertices $\Gamma^\Lambda$ (effective interactions) carry multiple indices: $i_1$ and $i_2$ for lattice sites whereas $\omega_1', \omega_2'$ and $\omega_1, \omega_2$ are frequencies for the pair of sites before and after the interaction. Contributions from higher order vertices are approximated by the Katanin term [38].

Care must be exercised to efficiently parametrize the vertices in order to render the numerical task tractable. In particular, the SS model has non-symorphic lattice symmetry, with four sites per unit cell shown in colors $\alpha = r, g, b, y$ in Fig. 1 and no $C_4$ symmetry as in the $J_1$-$J_2$ model. To avoid using color indices in $\Gamma^\Lambda_{ij}, \omega$, we pick a $\alpha = r$ site located at the origin as $i_1$. Other vertices for sites of different color $\alpha = g, b, y$ can be obtained from the central $r$-site with appropriate rotation and lattice translation [39]. We retain all $i_2$ within a radius $|r_{i_2} - r_{i_1}| < R_{\text{max}}$ in $\Gamma^\Lambda_{ij}$ and emphasize that the FRG equations describe infinite systems without a boundary. Here $R_{\text{max}}$ merely places an upper cutoff for the correlations retained in the numerics. As to the frequency variables, we rewrite $\Gamma^\Lambda(\omega_1', \omega_2'; \omega_1, \omega_2)$ as functions of the Mandelstam variables $s, t$ and $u$ [18] which manifestly enforce the frequency conservation. Finally, we discretize the frequency using a logarithmic mesh of $N_\omega$ points extending from the ultraviolet scale $\Lambda_{\text{UV}} = 10^2 J_2$ to the infrared scale $\Lambda_{\text{IR}} = 10^{-2} J_2$. Typically, $N_\omega = 48$ provides good frequency resolution, and further increasing $N_\omega$ will not alter the results appreciably. We take $R_{\text{max}} = 10$ which amounts to $N_L = 441$ lattice sites within the correlation radius. In total, this gives a coupled system of $N_L \times N_\omega^2 \sim 50$ million running couplings.

**Correlation functions and susceptibilities.**—To detect the emergence of long range order as $\Lambda \to 0$, correlation functions at each renormalization scale can be obtained from the $\Sigma^\Lambda$ and $\Gamma^\Lambda$ via standard calculations involving Feynman di-
agrams. For example, the spin-spin correlation function is given by
\[
\chi_{ij}(\omega) = \int_0^\infty d\tau e^{i\omega\tau} \langle TS^z_i(\tau)S^z_j(0) \rangle
\]
\[
= S^z_i \langle S^z_j + S^z_j \rangle , \quad (2)
\]
where black dots represent the spin matrix $S^z_i = \sigma^z_i/2$, filled square represents vertex $\Gamma^\Lambda$ and lines with arrows are dressed Green functions that contain the self-energy. The scale dependence of $\chi$ is suppressed for brevity. We find that it is necessary to distinguish the flows of spin correlations for different bonds, i.e. pairs of $(i, j)$, because the symmetry breaking patterns in the SS model are rather complex and involving valence bond orders. For a given site $i$ of color $\alpha$, one can find $\tilde{X}_\alpha(p)$, the Fourier transform of Eq. (2) in the limit of $\omega \to 0$. It is also convenient to define spin susceptibility
\[
\chi(p) = \lim_{\omega \to 0} \frac{1}{4} \sum_\alpha \sum_j e^{i(p,\pi)\cdot r_j} \chi_{ij}(\omega) = \frac{1}{4} \sum_\alpha \tilde{X}_\alpha(p) , \quad (3)
\]
where the $\alpha$ sum is over the four sites of different colors within the unit cell, the $j$ sum is over all sites, and the limit $\omega \to 0$ is taken in the end. The spin susceptibility defined in Eq. (3) has no bond resolution, but its divergence (or lack thereof) and its profile in momentum space offer a quick diagnosis of the incipient orders as the ratio $J_1/J_2$ is changed. Finally, we define a set of plaquette susceptibilities to detect the plaquette valence bond order. They measure the bond-resolved responses, e.g. the change in $\chi_{ij}$, due to a small bond modulation
\[
\chi_{ij}^p = -\frac{1}{J_1} \frac{\partial \chi_{ij}}{\partial \kappa} \bigg|_{\kappa \to 1} = -\frac{\partial \chi_{ij}}{\partial (\delta J_1)} \bigg|_{\delta J_1 \to 0} \quad (4)
\]
with $J_1$ and $J_2$ fixed. A dramatic enhancement of the $\chi_{ij}^p$ around the shaded plaquettes indicates an instability against a small fluctuation of modulation $\delta J_1$. To compute $\chi_{ij}^p$, we perform two runs of FRG flow with bare couplings $(\kappa J_1 = J_1 + \delta J_1, J_1, J_2)$ and $(J_1, J_1, J_2)$ for a given bond $(i, j)$. The procedure is expensive but provides invaluable insights.

Phase diagram.— The final results of our FRG calculation are summarized in the phase diagram shown in Fig. 1(b). It contains four phases as $J_1/J_2$ is varied at fixed $\kappa = 1$. For each phase, a representative value of $J_1/J_2$ is chosen to illustrate its characteristic FRG flow pattern in two complementary ways. First is the momentum distribution $\tilde{X}_\alpha(p)$ near the end of the flow (insets), where the peak momenta are labelled by white ‘x’ in the extended Brillouin zone [40]. Next is the flow of spin susceptibility $\chi(p)$ with the RG scale $\Lambda$ (main panels) for different channels, i.e., different values of $p$. For example, the channel $p = (\pi, 0)$ is shown in blue, the $(\pi, \pi)$ channel in orange, while the flow for the peak momenta labelled by ‘x’ is in red. Clearly, the leading channels for the four phases are distinct. Take the case $J_1/J_2 = 1.0$ for example, from the inset it is clear that $\tilde{X}_\alpha(p)$ is peaked at $p = (\pi, \pi)$. Accordingly, the FRG flow for $\chi(\pi, \pi)$ (in orange) is most dominant and rises rapidly as $\Lambda$ is reduced. The flow breaks down around $\Lambda^* \approx 0.2$, signaling a physical divergence and the onset of magnetic long-range order as seen in many FRG calculations. Thus the Neel phase can be identified unambiguously from the $(\pi, \pi)$ peak and the flow divergence.

Outside the Neel phase, the flows appear smooth down to the lowest $\Lambda$. This is perhaps not that surprising because spin rotational symmetry is not broken in the dimer or plaquette phase. Yet by inspecting the two cases $J_1/J_2 = 0.3$ and 0.7 in Fig. 1, it is apparent that their spin correlations are rather different, e.g., they have different peak momenta or leading channels. Unfortunately, the information contained in $\chi(p)$ or $\tilde{X}_\alpha(p)$ is too crude to differentiate the dimer from the plaquette phase. In what follows, we show that this can be achieved by the FRG flow of bond-resolved spin correlation $\chi_{ij}$.

Dimer phase as a fixed point.— Fig. 2(a) compares the flows of $\chi_{ij}$ for the diagonal bond (red lines in the inset) at different values of $J_1/J_2$. One notices a remarkable phenomenon: for all $J_1/J_2 < 0.6$, they flow to the same exact value $\approx 0.41$ in the infrared $\Lambda \to 0$. This renormalization group fixed point defines a robust phase with constant spin correlation along the diagonal. This is nothing but the dimer phase, in accordance with the known fact that the ground state wave function in this region is a product state of isolated spin singlets, frozen with respect to $J_1/J_2$ with constant energy up to a critical point. To determine its phase boundary, Fig. 2(b) plots the diagonal bond correlation in the infrared limit versus $J_1/J_2$. It stays completely flat before dropping rapidly in a linear fashion. Linear regression (black lines) yields an intersection point at $J_1/J_2 = 0.67$ which we take as the estimated phase transition point. This critical value is impressively close to 0.675 from large-scale DMRG [3]. The agreement provides strong
FIG. 3. Identification of the phase boundaries from bond-resolved plaquette susceptibility $\chi^P_{ij}$, which measures the response of a given bond $(i, j)$ [red lines in insets] to a small increase $\delta J_1$ in the shaded squares. (a) A pronounced peak of $\chi^P_{ij}$ around $J_1/J_2 = 0.67$ signals the onset of plaquette order. (b) The suppression dip of $\chi^P_{ij}$ at 0.77 indicates the plaquette order is superseded by a new phase, the spin liquid with distinct inter-plaquette correlations. (c) An example of longer range $\chi^P_{ij}$ that also develops peak/dip at the two critical points above. (d) The momentum space degeneracy $\eta$ of $\tilde{\chi}^P(p)$ is peaked at $J_1/J_2 \approx 0.82$, which marks the transition into the Neel phase.

Consistency of our FRG and the advantage of introducing the quantity $\chi^P_{ij}$. It is not a divergence because higher-order vertices are truncated in the current implementation. The onset of plaquette order also manifests in longer range inter-plaquette correlations. Fig. 3(c) depicts the $\chi^P_{ij}$ for a bond between an $r$-site and a $b$-site from two shaded squares along the lattice diagonal. It too has an enhancement peak at $J_1/J_2 \approx 0.67$.

Further analysis of $\chi^P_{ij}$ also points to the demise of the plaquette phase. A pristine plaquette order is adiabatically connected to the limit of decoupled plaquette singlets (shaded squares in Fig. 1 without red or dashed bonds). Upon increasing $J_1/J_2$, the plaquette order eventually yields to a state with homogeneous bond energies and very different spin correlations. One possibility is a liquid state where the shaded and empty squares are entangled to feature strong inter-plaquette correlations. The change in correlation is apparent in Fig. 3(c): after the peak, $\chi^P_{ij}$ changes sign to develop a sharp dip at $J_1/J_2 \approx 0.78$, suggesting the onset of a new phase. This interpretation is supported by the plaquette susceptibility $\chi^P_{ij}$ shown in Fig. 3(b). It measures the change to the bonds around the empty plaquettes in response to $\delta J_1 > 0$ in the nearby shaded squares. When $J_1/J_2$ is reduced from above toward 0.77, a small $\delta J_1$ leads to significant weakening of the antiferromagnetic bonds (red lines) around the empty squares, i.e. decoupling of the shaded plaquettes to break translational symmetry. Thus the pronounce dip of $\chi^P_{ij}$ at $J_1/J_2 \approx 0.77$ marks the upper critical point of the plaquette phase.

At the very least, the dramatic variations of $\chi^P_{ij}$ are at odds with the scenario that the plaquette phase persists after $J_1/J_2 \approx 0.77$.

A sliver of spin liquid.—The existence of a novel phase after $J_1/J_2 \approx 0.77$ can be inferred independently from the spin correlation $\chi_{ij}$ for the diagonal bond shown in Fig. 2(b) (dashed lines). Here it becomes flat, i.e. independent of $J_1/J_2$, in the infrared limit. The behavior is distinct from that of a plaquette valence bond solid or a Neel antiferromagnet, for which $\chi_{ij}$ increases with $J_1$. Since the spin susceptibility flow is continuous down to $\Lambda \rightarrow 0$ as shown in Fig. 1(b), the only plausible scenario seems to be that this FRG fixed point corresponds to a liquid phase. With further increase in $J_1$, the flat top of diagonal $\chi_{ij}$ is terminated by an upturn around $J_1/J_2 \sim 0.82$, signaling another phase transition. To precisely locate the onset of the Neel order, we adopt an independent criterion [43]. In the postulated spin liquid region, the spin susceptibility $\tilde{\chi}^S(p)$ develops broad maxima, instead of a sharp peak, in momentum space, see the case of $J_1/J_2 = 0.8$ in Fig. 1(b). We can quantify the peak degeneracy by $\eta$, the percentage of $p$ points with $\tilde{\chi}^S(p) \geq 0.9\max_\delta[\tilde{\chi}^S(p)]$. A similar method was employed in [44] for a different system. The result is shown in Fig. 3(d). As the Neel phase is approached, the broad maxima coalesce into a sharp peak at $(\pi, \pi)$, after which $\eta$ drops quickly. The peak location of degeneracy $\eta$ at $J_1/J_2 = 0.82$ serves as an accurate estimation for the transition from the spin liquid to the Neel phase, in excellent agreement with the phase boundary obtained from large scale DMRG [4].

Conclusions.—Our high-resolution FRG analysis of the SS model identifies four phases separated by three critical points

evidence for the validity and accuracy of our FRG calculation.

Plaquette valence bond solid.—Identification of plaquette order from FRG has been an open challenge. In earlier studies, a plaquette susceptibility $\chi^{PV}_P$ was defined as the propensity towards translational symmetry breaking with respect to a small bond modulation bias [18, 25, 33]. Enhancement of $\chi^{PV}_P$ has been reported, but to our best knowledge, plaquette order has not been positively identified using pseudofermion FRG so far. For the generalized SS model, we have confirmed that $\chi^{PV}_P$ is indeed enhanced within a broad region stretching from $J_1/J_2 \sim 0.5$ to 0.7 when compared to its values within the Neel phase (see [39] for details). But it only exhibits a smooth crossover with $J_1/J_2$ due to the lack of bond resolution. This has motivated us to introduce a more refined measure, the bond-resolved plaquette susceptibility $\chi^P_{ij}$ in Eq. (4).

Fig. 3(a) illustrates the $\chi^P_{ij}$ for the horizontal/vertical bonds within the slightly strengthened plaquettes (shaded squares), denoted by $\chi^P_{ij}$. While it is more or less featureless at the ultraviolet scale, as RG steps are taken and $\Lambda$ is reduced, $\chi^P_{ij}$ gains nontrivial dependence on $J_1/J_2$. In particular, in the infrared limit $\chi^P_{ij}$ develops a pronounced peak around $J_1/J_2 \approx 0.67$ [41]. The dramatic enhancement of plaquette susceptibility marks the onset of plaquette order. This independent diagnosis of the dimer-to-plaquette transition agrees very well with the linear regression result above, showing the self-
summarized in Fig. 1. Key technical insights are retrieved by monitoring the renormalization group flows of bond-resolved spin-spin correlation functions and susceptibilities. The good agreement with other established numerical methods on the locations of the phase boundaries attests to the accuracy of FRG which takes into account quantum fluctuations in all the channels without bias by tracking millions of effective couplings at each scale $\Lambda$. The implementation and analysis strategies reported here can be applied to study other quantum spin Hamiltonians with unconventional magnetic orders using pseudofermion FRG. In particular, our result supports the existence of a finite spin liquid phase rather than a deconfined quantum critical point between the plaquette and Neel phase. It motivates future theoretical work to further elucidate the nature and extent of this phase, and precision measurements to locate and probe spin liquid in SrCu$_2$(BO$_3$)$_2$.

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ahmetkeles99@gmail.com

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[38] See Supplemental Material at [URL will be inserted by publisher] for implementation details of Pseudofermion FRG and discussions with additional data which include the additional references [45–47].

[39] Typically $\chi(p)$ is plotted, which is obtained by summing over $\chi_\alpha$ according to Eq. (3) to obey all crystal symmetries within the Brillouin zone. Here we choose to show $\chi_r$ because it appears less cluttered.

[40] We have checked that the location of the peak remains the same upon increasing the correlation cutoff $R_{max}$, adopting a finer frequency grid with lower $\Lambda_{IR}$ and taking smaller bias $\Delta J_1$.

[41] We note that the suppression dips in Fig. 3(b) and Fig. 3(c) do not coincide exactly. This discrepancy is expected to reduce with the inclusion of higher order vertices.

[42] The Neel order is rather weak for $J_1/J_2 < 0.9$, so it is numerically hard to pinpoint at what $J_1/J_2$ value exactly the divergence sets in. It is more accurate to consider the degeneracy of susceptibility as defined in the main text.

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In the supplementary materials, we give technical details about the implementation of lattice symmetries in the two-particle irreducible vertex. We present a classical phase diagram of generalized Shastry-Sutherland model by taking the static limit of the vertex. We give a detailed scan of the phase diagram and show flow of susceptibilities in the momentum space. We show the plaquette susceptibility flow as defined in the previous FRG studies and further motivate our bond-resolved susceptibility calculations.

1. Lattice Symmetries in Shastry-Sutherland Model

In the Shastry-Sutherland model, there are four lattice sites per unit cell which requires four different sites with colors \( \alpha = r, g, b, y \) as reference site. To avoid additional numerical burden stemming from such sublattice symmetries we consider the \( \alpha = r \) site at the origin \( i_0 \) and consider a cluster of sites with index \( i_2 \) within the radius \( |r_i - r_{i_0}| < R_{\text{max}} \) independent of sublattice type at \( i_2 \). In the flow equations and susceptibility calculations, whenever we need vertex functions of the form \( \Gamma_{j_2} (s, t, u) \) with a site \( j \) of different color \( \alpha = g, b, y \), which is not the sublattice type of the central reference site, we use the following lattice symmetry transformations:

\[
\Gamma_{j_2} (s, t, u) \quad \text{where} \quad j' = \begin{cases} \ (\delta x, \delta y) & \text{if } j \text{ is } r \\ \ (\delta y, -\delta x) & \text{if } j \text{ is } g \\ \ (-\delta x, -\delta y) & \text{if } j \text{ is } b \\ \ (-\delta y, \delta x) & \text{if } j \text{ is } y \\ \end{cases}
\]  

(S1)

where \( (\delta x, \delta y) = r_{i_2} - r_j = (x_{i_2} - x_j, y_{i_2} - y_j) \). Furthermore, the vertex functions have the following symmetries

\[
\Gamma_{i_2} (s, t, u) = \begin{cases} \ Gamma_{i, s_2} & \text{if } i_2 \text{ is } r \\ \ Gamma_{i, g} & \text{if } i_2 \text{ is } g \\ \ Gamma_{i, b} & \text{if } i_2 \text{ is } b \\ \ Gamma_{i, y} & \text{if } i_2 \text{ is } y \\ \end{cases}
\]  

(S2)

which is used to set up a frequency grid only for positive \( s, t \) and \( u \) which reduces the computational cost significantly. Exchange of the site indices \( i_1 \) and \( i_2 \) also has to be done in a way to respect the sublattice symmetry. In the vertex \( \Gamma_{i_2} \), we assume \( i_1 \) is the reference site at the origin, \( i_2 \) is a site with coordinates \((x, y)\) then the site flipping operation is done by \( \Gamma_{i_2} \rightarrow \Gamma_{i, k} \equiv \Gamma_{i, k} \) where \( k \) is the index of the site

\[
\Gamma_{i, k} (s, t, u) = \Gamma_{i_2} (s, t, u) \quad \text{where} \quad i_2 = \begin{cases} \ (-x, -y) & \text{if } i_2 \text{ is } r \\ \ (-y, x) & \text{if } i_2 \text{ is } g \\ \ (x, y) & \text{if } i_2 \text{ is } b \\ \ (y, -x) & \text{if } i_2 \text{ is } y \\ \end{cases}
\]  

(S3)

for \( r_{i_2} = (x, y) \) and \( r_{i_0} = (0, 0) \).

Fourier transform with sublattice degrees

\[
\chi(\omega, k) = \frac{1}{4} \sum_{\alpha=\{r, g, b, y\}} \sum_{j} e^{i\mathbf{k}\cdot(r_j-r_{i_0})} \chi_{ij}(\omega) = \frac{1}{4} \left[ \chi_r(\omega, k_x, k_y) + \chi_g(\omega, k_x, -k_y) + \chi_b(\omega, -k_x, -k_y) + \chi_y(\omega, -k_x, k_y) \right]
\]  

(S4)

where we defined the susceptibility contribution from the central site in the unit cell

\[
\chi_{ij}(\omega) = \sum_{j} e^{-i\mathbf{k}\cdot r_j} \chi_{ij}(\omega)
\]  

(S5)

and the lattice is setup with \( r_{i_0} = (0, 0) \). The last equality in Eq. S4 can be proven using the symmetry relations given in Eq. S1 and S3.
This shows the power of pfFRG even in this simplified calculation. Similar calculation for generalized SS model with finite $\kappa$ is shown in the right panel of Fig. S1.

2. STATIC VERTEX LIMIT: CLASSICAL PHASE DIAGRAM

Calculation of the flow equations for frequency independent vertices gives the classical phases of the system and corresponds to Random Phase Approximation (RPA) known in the study of correlated electrons. Here, we include results of our calculation in static limit, which gives a good starting point for understanding the phase diagram.

In the limit of static vertex, self energy vanishes and scale dependent propagator becomes

$$G^\Lambda(\omega) = \frac{\Theta(|\omega| - \Lambda)}{i\omega + i0^+ \text{sign}(\omega)}$$  \hspace{1cm} (S6)

where we have added an infinitesimal part in the denominator. Flow equation for the vertex becomes

$$\frac{d}{d\Lambda} \Gamma_{sij} = 2 \pi \Lambda^2 \left[ \sum_j \Gamma_{sif} \Gamma_{sjf} - 2 \Gamma_{sii}^2 + \Gamma_{sij} \Gamma_{sji} \right]$$  \hspace{1cm} (S7)

The first term in the susceptibility diagram in the main text can be calculated as

$$\delta_{ij} \text{Tr} \left[ \frac{1}{2} \sigma^z \frac{1}{2} \sigma^z \right] \int_{-\infty}^{\infty} d\omega \frac{1}{2\pi} G^\Lambda(\omega) G^\Lambda(\omega) = \delta_{ij} \frac{1}{2} \int_{-\infty}^{\infty} d\omega \frac{1}{(i\omega + i0^+)^2} = \delta_{ij} \frac{1}{2\pi(\Lambda + 0^+)}$$  \hspace{1cm} (S8)

The second term with the static vertex can be calculated similarly giving

$$\chi_{ij}^\Lambda = \frac{1}{2\pi \Lambda} \frac{1}{\pi^2 \Lambda^2} \Gamma_{sij}^\Lambda$$  \hspace{1cm} (S9)

In the numerical solution of flow equation Eq. S7 in static limit, vertex functions tend to diverge at small energies signaling the onset of classical long-range orders. In our numerics, when any value in $\Gamma_{ij}$ reaches a maximum value $\Gamma_{\text{max}}$, we stop the flow and identify the corresponding scale as the critical scale $\Lambda_c$.

The critical scale $\Lambda_c$ found from static FRG is shown in Fig. S1. For small $J_1$, the variation of $\Lambda_c$ with $J_1$ is very small. As we approach the phase transition around $J_1 = 0.5$, $\Lambda_c$ start to deviate from this base value somewhat quadratically and finally it shows linear increase with $J_1$ inside the known Neel phase. Then, the first derivative of the transition scale $\Lambda_c$ with respect to $J_1$ is well approximated with a step-like function. To determine the phase transition point, we calculate the critical scale $\Lambda_c$ for different values of $J_1$ and calculate its first derivative $d\Lambda_c/dJ_1$ numerically and fit the result to a function $a + b \tanh[c(x - x_0)]$.

The value for $x_0$ obtained from this fitting with give approximation for the transition point within the static FRG. We determine the kink position $J_c \approx 0.71$, is the midpoint of the two phase transition points reported in the state-of-the-art DRMG simulations. This shows the power of pfFRG even in this simplified calculation. Similar calculation for generalized SS model with finite $\kappa$ is shown in the right panel of Fig. S1.
3. DETAILED SUSCEPTIBILITY FLOWS IN REAL AND MOMENTUM SPACE

Susceptibility flows in momentum space and real space are shown in Fig. S2 with more data points along the phase diagram.

4. PLAQUETTE SUSCEPTIBILITY WITHOUT BOND RESOLUTION

We probe the plaquette correlations based on the methodology used in the previous pFRG studies\(^1,5\) for comparison. We strengthen the couplings between the spins in the shaded plaquette (bond towards upper and right nearest neighbors in Fig. 1 in the main text) \(J_1 \to J_1 + \delta J_1\) and weaken the couplings between the spins in the empty plaquette (lower and left neighbors in Fig.1) \(J_1 \to J_1 - \delta J_1\) in the initial conditions by an infinitesimal amount \(\delta J_1\) for fixed \(J_2 = 1\) and calculate the following quantity

\[
\chi_{\text{PVB}} = \frac{J_1 \chi_{ij\neq P} - \chi_{ijP}}{\delta J_1 \chi_{ij\neq P} + \chi_{ijP}}
\]

where \(\chi_{ij\neq P}\) and \(\chi_{ijP}\) are susceptibilities of the bonds in the strengthened and weakened plaquettes, respectively.

Calculation of this quantity from full FRG is shown in the following Fig. S3. Note that there is no bond resolution in \(\chi_{\text{PVB}}\).

We observe that it increases about five times around \(J_1 = 0.6 - 0.7\), which is in the middle of the putative plaquette phase, and goes back down as we approach to Neel phase. In the lower panel, we plot \(\chi_{\text{PVB}}\) along a denser scan of the phase diagram at different RG scales.

\(\chi_{\text{PVB}}\) defined above is able to discern short-ranged plaquette phase from the Neel successfully but it seems to distinguish dimer and plaquette phases only weakly. It can be seen that this susceptibility is significant even for \(J_1 < 0.5\), which was shown to be the dimer phase unambiguously in the main text.

5. DEFINITION OF BOND-RESOLVED PLAQUETTE SUSCEPTIBILITY

One can motivate the alternative bond-resolved susceptibility by considering the expression \(\langle S_i S_j \rangle = \text{Tr}(S_i \cdot S_j e^{-\beta H})/\text{Tr}(e^{-\beta H})\) and taking the derivative of both sides with respect to \(J_{il}\) (coupling between spins \(S_k\) and \(S_l\)) in a way that \(i, j, k, l\) are corners of a given plaquette pattern while keeping all other couplings fixed. It is easy to show \(\partial \chi_{ij}/\partial J_{il} = -\beta (\langle S_i \cdot S_j \rangle \langle S_k \cdot S_l \rangle) + \beta (\langle S_i \cdot S_j \rangle \langle S_k \cdot S_l \rangle)\) which is the connected average of the four spin operators at sites \(i, j, k, l\).

To calculate this equation in practice, we start with Shastry-Sutherland model for a given nearest-neighbors coupling \(J_1\) by taking \(J_2 = 1\), we calculate the susceptibility \(\chi_{ij}[J_1]\) as usual; we increase the strength of exchange coupling \(J_1 \to J_1 + \delta J_1\) only for the target plaquette (upper neighbor and right neighbor couplings \(J_1 + \delta J_1\) but lower neighbor and left neighbor couplings
are still $J_1$) and calculate the resulting susceptibility $\chi_{ij}[J_1 + \delta J_1]$; finally define the following correlation function

$$\chi_{ij}^P \equiv \frac{\chi_{ij}[J_1 + \delta J_1] - \chi_{ij}[J_1]}{\delta J_1}.$$ (S11)

This procedure requires running numerically expensive pfFRG simulation for two points that are infinitesimally close to each other in the phase diagram but it provides a useful perspective.

6. HIGHER-LOOP CONTRIBUTIONS

In this section, we discuss the limitation of our calculation and the possible extension of our work to include higher-loop contributions. To solve the formally exact flow equations, most FRG implementations involve two numerical aspects, both of which influence the accuracy and the computational cost of the algorithm. First is the truncation of the infinite hierarchy of flow equations to a certain loop order plus higher order correction terms. In the context of pseudo-fermion FRG, the one-loop truncation plus the Katanin terms as implemented here has been extensively applied and benchmarked. For interacting system, no numerical implementation of FRG manages to include all diagrams to infinite loop order. Empirically, most known divergences (instabilities toward long-range order), and the mutual influence of these instabilities, seem to be accounted already at the level of one-loop diagrams for interacting fermions. Second is the resolution for momentum and frequency in the numerical solution of the truncated flow equations. The term “high-resolution” throughout our manuscript refers to the frequency as well as the momentum resolution (or equivalently, the site resolution, which in our implementation is determined by the cut-off correlation length in real space). A main technical effort of our work is to achieve sufficient frequency and site resolution, which as we have seen in the main text is crucial to analyze the plaquette susceptibility, leading to the identification of an unexpected spin liquid region. For example, with 64 grid points in the frequency and $21 \times 21 = 441$ lattice sites, we end up with 120 million running couplings. Decreasing the frequency resolution from 64 to 48 decreases to the number of coupling to about 50 million but the change in the flow of spin susceptibilities are almost the same at the level of machine precision. The computation is made feasible by migrating to GPU. The improved resolution not only can help identify complex long-range orders hitherto impossible, but also can provide fresh physical insight by comparing the various bond-resolved susceptibilities.

A natural question is then “how important are additional higher loop contributions?” A definitive answer to this questions would require solving the flow equation with additional higher-loop terms systematically taken into account, which is beyond the computing resources available to us at the moment. But in principle, bond-resolved spin and plaquette susceptibilities can be calculated with additional higher-loop contributions. This would be the subject of a future research.

Very recently, a few groups have begun to shed light on this open question by explicitly taking into account additional higher-loop contributions\(^6,7\). In assessing these results and discussing higher-loop expansions, one must carefully distinguish FRG for itinerant fermions and pf-fRG for quantum spin systems, because the nature of the loop expansion, hence the role played by the higher loops are drastically different. For itinerant fermions, such as the one-band Fermi-Hubbard model on square lattice studied by Hille et al.\(^8\), one motivation to include higher loop contributions is to accurately describe the momentum and frequency dependence of the self-energy, which was neglected in many of the earlier N-patch FRG studies. The other motivation
is to achieve quantitative agreement with Parquet approximation and Determinant quantum Monte Carlo. It remains to be seen how such “improved fRG*” performs when away from half-filling.

For quantum spin systems, two recent works investigated the higher-loop corrections in pf-fRG⁶,⁷. As we outlined in the main text, the pseudo-fermion representation of spin models leads to a strongly interacting system where the bare kinetic energy is zero. Here the loop expansion is motivated more by the spirit of 1/S or 1/N expansion⁸,¹⁰. (In pseudo-fermion FRG, the frequency dependence of self-energy plays a crucial role and is already taken into account from the very beginning.) To our knowledge, there still lacks a satisfactory understanding of the role of higher-loop contributions, including its convergence properties when long-range or spin liquid phases are approached in the infrared limit. The state-of-the-art multiple-loop pf-fRG seems to imply that the divergence towards long-range order, or the lack thereof, is dictated by the leading contributions contained in the level-one truncation (one loop plus Katanin terms, as implemented in our work). Moreover, our confidence in the performance of pf-fRG applied to the Shastry-Sutherland (SS) model stems from its (unexpected) remarkable agreement with other independent, state-of-the-art methods such as DRMG regarding the phase boundaries. Future work will elucidate whether or how the predicted phase boundary may change with higher order contributions included in a consistent manner.

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