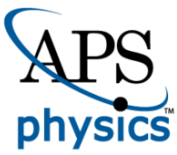

Dynamics and Instabilities of the Shastry-Sutherland Model

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Dynamics and Instabilities of the Shastry-Sutherland Model

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We study the excitation spectrum in the dimer phase of the Shastry-Sutherland model by using an unbiased variational method that works in the thermodynamic limit. The method outputs dynamical correlation functions in all possible channels. This output is exploited to identify the order parameters with highest susceptibility (single or multi-triplon condensation in a specific channel) upon approaching a quantum phase transition in the magnetic field vs. J'/J phase diagram. We find four different instabilities: antiferro spin nematic, plaquette spin nematic, stripe magnetic order and plaquette order, two of which have been reported in previous studies.

PACS numbers:

The Shastry-Sutherland model (SSM) has become a paradigmatic Hamiltonian of frustrated quantum magnetism [1, 2] because it includes an exactly solvable ground state [1], very heavy low-energy excitations [3–8], exotic phases obtained upon varying the ratio J'/J between two competing exchange constants [4, 7, 9–16], and series of magnetic field induced magnetization plateaux [3, 17–31]. Its realization in $SrCu_2(BO_3)_2$ [3, 32, 33] enabled various experimental studies, including magnetization [32, 34–39], specific heat [40], inelastic neutron scattering (NS) [41–47], far-infrared [48], electron spin resonance (ESR) [49, 50], Raman scattering [51] and nuclear magnetic resonance (NMR) [37, 52, 53]. These studies revealed that a finite Dzyaloshinskii–Moriya (DM) interaction [54, 55] must be added to the SSM in order to account for several proerties of $SrCu_2(BO_3)_2$ [42–44, 46, 49, 50, 53, 56–60].

Despite the great theoretical efforts devoted to the SSM, the problem is still far from being solved. Perturbative approaches are only applicable in narrow regimes and conventional numerical methods suffer from severe size effects. As a consequence, the nature of the quantum phase diagram has been debated for a long time [4, 7, 9–16]. It is thus desirable to develop and apply alternative approaches. The infinite projected entangled-pair states (iPEPS) is an example of an alternative approach that works in the thermodynamic limit [16, 29, 61]. However, it relies heavily on the initial guess of the physical states and it is difficult to extract dynamical responses.

In this Letter, we introduce an *unbiased* variational method to calculate the excitation spectrum and dynamical responses (susceptibilities) of the SSM in the dimer phase [62]. The method works in the thermodynamic limit and it complements alternative approaches like iPEPS. The basic idea was originally introduced to compute the single-hole dispersion of the square lattice $t-J$ model [63, 64]. The same method was applied to the Shastry-Sutherland lattice $t-J$ model [65, 66]. Here we exploit this idea to compute dynamical correlators and dominant instabilities. By working in a reduced Hilbert space, *which preserves all model symmetries*, we obtain low energy excitations classified by quantum numbers. We then predict the character of the neighboring phases by detecting the order parameter with highest susceptibility. Besides confirming the previously reported plaquette order and antiferro spin nematic phases, we find two new phases, namely a plaquette spin nematic phase and stripe magnetic ordering, induced by simultaneously increasing the magnetic field and J'/J. In particular, the plaquette spin nematic phase explains the nature of the two-triplon states (pinwheels) that crystallize at higher field values [29].

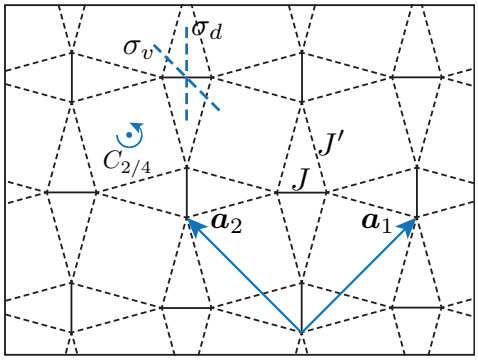

Figure 1. Lattice structure of the SSM. Intra-dimer and inter-dimer exchanges are denoted by J(solid line) and J'(dashed line). The basis of the lattice is labeled by $\{\boldsymbol{a}_1, \boldsymbol{a}_2\}$. The point group operations $\{\sigma_v, \sigma_d, C_2, C_4\}$ are denoted accordingly.

We consider the spin-$\frac{1}{2}$ SSM under a magnetic field [1]:

$$\mathcal{H} = J \sum_{\langle ij \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J' \sum_{\langle\langle ij \rangle\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - h \sum_i S_i^z, \qquad (1)$$

where $\langle ij \rangle$ and $\langle\langle ij \rangle\rangle$ denote intra-dimer and inter-dimer neighbors. The unit cell has 4 sites (see Fig. 1). The exact ground state for small enough J'/J and h is a direct product of singlet states on all dimers [1]. The elementary excitations of this "dimer phase" are singlet-triplet excitations known as triplons. Triplons are dressed by quantum fluctuations with a magnetic correlation length ξ that increases with J'/J. Methods that can account for the spatial extent of these quantum fluctuations should provide a good description of the low-energy excitation spectrum of the dimer phase.

We start the process by creating local excited states $|\varphi_i\rangle$ (e.g., single and two triplons). We then project these representative states into subspaces with fixed momentum $\boldsymbol{k}$,

$$|\varphi_i(\boldsymbol{k})\rangle \equiv \frac{\hat{P}_{\boldsymbol{k}}|\varphi_i\rangle}{\sqrt{\langle\varphi_i|\hat{P}_{\boldsymbol{k}}|\varphi_i\rangle}}, \tag{2}$$

where the projector is defined as $\hat{P}_{\boldsymbol{k}} \equiv \frac{1}{N}\sum_{\boldsymbol{r}} e^{i\boldsymbol{k}\cdot\boldsymbol{r}}\hat{T}(\boldsymbol{r})$. $N \to \infty$ (thermodynamic limit) is the total number of unit cells, and $\hat{T}(\boldsymbol{r})$ is the translation operator. Application of $\mathcal{H}$ to $|\varphi_i(\boldsymbol{k})\rangle$ generates new states that dress the corresponding quasi-particle excitation. This procedure can be applied iteratively to systematically improve the variational space. After M iterations, we obtain a basis $\{|\varphi_i(\boldsymbol{k})\}$ with good quantum numbers $\boldsymbol{k}$ and S^z_{tot}. The number of iterations determines the spatial range l of the fluctuations that dress the quasi-particle, so the method is then expected to produce accurate results for $l \gtrsim \xi$.

The eigenvalues and eigenvectors of the Hamiltonian restricted to the variational space are obtained by applying the Implicitly Restarted Arnoldi Method [67, 68]. The eigenvectors are classified by the Little Group of C_{4v} for each momentum $\boldsymbol{k}$. *A continuous phase transition* manifests via a vanishing gap (condensation) that signals a phase transition into a broken symmetry state. The symmetry of the new state is determined by the irreducible representation (IREP) of the eigenstate that becomes gapless. *To keep the method unbiased, the initial basis must not break the point group symmetry of* $\mathcal{H}$ [69].

For illustration, we first focus on the $S^z_{\text{tot}} = 0$ sector relevant to $h = 0$. We include $\mathcal{D} = 14$ $S^z_{\text{tot}} = 0$ initial states to start the iteration [70] and then apply Eq. (1) onto this basis to systematically increase the variational space [71]. After obtaining the lowest energy eigenstates, we use the eigenfunction to calculate S_{tot} and its IREP [72].

In contrast to the result obtained with perturbative continuous unitary transformations (CUTs) [7], we find that the first instability as a function of J'/J (for $h = 0$) takes place in the $S_{\text{tot}} = 0$ channel with IREP A_2 [73]. Figure 2 shows the evolution of the gap as a function of M. Convergence is reached beyond $M = 3$ for $J'/J \lesssim 0.5$, but the increase of ξ slows down the convergence for larger J'/J. Although Eq. (1) does not conserve the triplon number, the state that condenses is adiabatically connected with the corresponding $S_{\text{tot}} = 0$ IREP A_2 pure two-triplon state in the $J'/J \to 0$ limit (see Fig. 2).

We can read out the critical value of J'/J when this state condenses. Figure 3a shows the evolution of the critical value as we increase M (circles). At $M = 8$, $(J'_c/J)^{(M=8)} \approx 0.702$. Previous tensor network based calculations [15, 16] showed that the transition is actually of first order and the transition point is at $J'_c/J = 0.675$ [16]. A susceptibility analysis, like the one presented here, is in general inadequate to detect first order transitions. However, it can still be used to detect the nature of order parameter *if the system still transitions into the broken symmetry state with highest susceptibility* [74]. Given that the first order transition takes place when this susceptibility is still finite, J'_c/J turns out to be smaller than the value at which the susceptibility becomes divergent. This observation explains

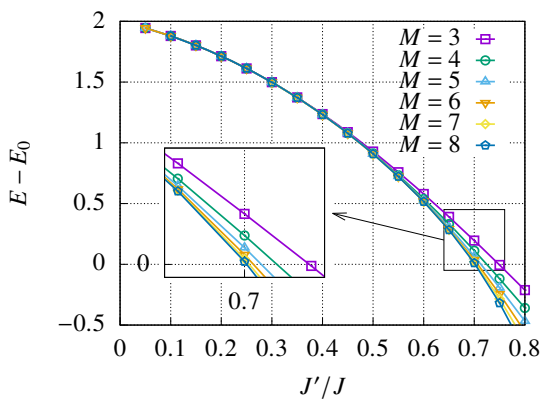

Figure 2. Gap of the lowest $S_{\text{tot}} = 0$, $\boldsymbol{k} = (0,0)$ A_2 state, at $h = 0$.

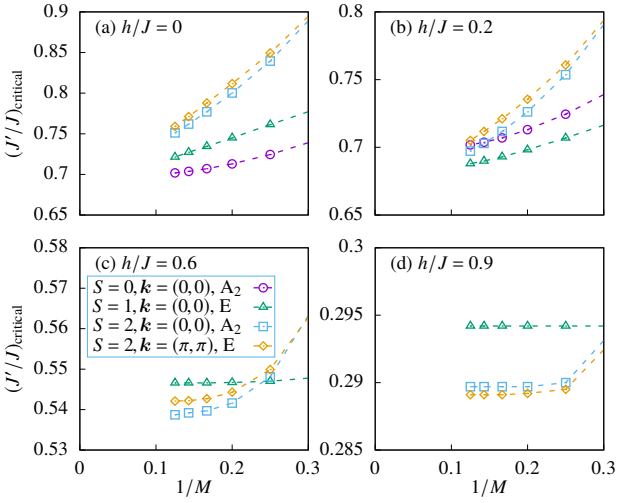

Figure 3. Critical values of J'/J for the condensation of different states at 4 different magnetic fields.

the difference between the values of J'_c/J obtained with both approaches. In addition, it illustrates their complementary nature. The unbiased susceptibility analysis can be used to detect candidates for broken symmetry states. These candidates can then be tested with biased variational techniques, such as iPEPS, which can produce more accurate values of the transition point.

Since the two-triplon bound state has $S_{\text{tot}} = 0$, the new ground state (condensate) must be non-magnetic. Furthermore, since the A_2 state is odd (even) under reflection (rotation) [70], the new ordered state should only break reflection but not rotation symmetries. These features are consistent with the previously reported plaquette ordering [10, 13, 15, 16]. Figure 4b shows a schematic plot of the corresponding bond ordering. As expected, $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle$ becomes different on different plaquettes and there is no magnetic order. In other words, the plaquette order parameter can be defined as $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j - \boldsymbol{S}_i \cdot \boldsymbol{S}_{j'} \rangle$, where ij and ij' are two bonds related by a mirror reflection (see Fig. 4b).

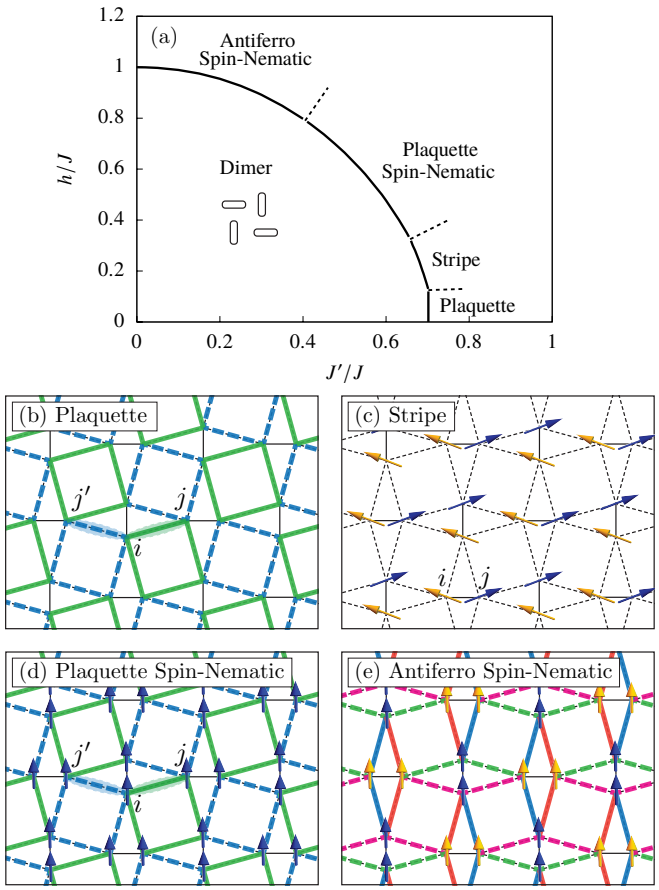

Figure 4. (a) Phase boundaries between the dimer state and its neighboring phases, obtained from $M = 8$ iterations. (b) Plaquette phase, order parameter defined as $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j - \boldsymbol{S}_i \cdot \boldsymbol{S}_{j'} \rangle$. (c) Stripe phase, order parameter defined as $\langle \boldsymbol{S}_i - \boldsymbol{S}_j \rangle$. (d) Plaquette spin-nematic phase, order parameter defined as $\langle S_i^+ S_j^+ - S_i^+ S_{j'}^+ \rangle$. (e) Antiferro spin-nematic phase (bond density wave), bonds with the same line (solid/dashed) but different colors have opposite $\langle S_i^+ S_j^+ \rangle$ while bonds with different lines have different $|\langle S_i^+ S_j^+ \rangle|$.

We consider now the case of non-zero magnetic field. The energy of excited states with finite $S^z_{\rm tot}$ decreases linearly in h. Figure 3b shows that the dominant instability for $h/J = 0.2$ corresponds to condensation of a state with $S_{\rm tot} = 1$, $\boldsymbol{k} = (0,0)$, and IREP E, leading to the stripe magnetic order depicted in Fig. 4c. The IREP E is a 2-dimensional representation corresponding to the two possible directions of the stripes (along $\boldsymbol{a}_1$ or $\boldsymbol{a}_2$). We note that the two same-color spins in the same unitcell are not identical (the corresponding mirror symmetry is broken).

The stripe state has the highest susceptibility over a narrow range $0.66 \lesssim J'/J \lesssim 0.70$ for $M = 8$ iterations (see Fig. 4a). Due to the frustrated exchange interactions, the energies of a few other states are not much higher than the stripe state [70]. Among them, the lowest one is a two-triplon state with $\boldsymbol{k} = (0,0)$ and IREP B_1, corresponding to vector chiral order [75–77]. Although their energies are slightly higher than the stripe magnetic instability within $M \leq 8$, the situation may change in the $M \to \infty$ limit, or if small perturbations are added to the original model.

The $S_{\rm tot} = 2$ excited states take over for higher magnetic field values. Figure 3c shows that for $h/J = 0.6$ the lowest excited state is the $S_{\rm tot} = 2$ two-triplon bound state with momentum $\boldsymbol{k} = (0,0)$ and IREP A_2. The fact that this state and the $S_{\rm tot} = 0$ state that condenses at zero field belong to the same point group IREP A_2 indicates that the condensation of the $S_{\rm tot} = 2$ A_2 state also leads to "plaquette" ordering (shown in Fig. 4d); the difference being that the $S_{\rm tot} = 2$ condensate also breaks the U(1) symmetry group of global spin rotations along the field direction, leading to spin-nematic ordering. In other words, the local bond order parameter is $\langle S_i^+ S_j^+ - S_i^+ S_{j'}^+ \rangle$ instead of $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j - \boldsymbol{S}_i \cdot \boldsymbol{S}_{j'} \rangle$ (ij and ij' denote two bonds connected by a mirror reflection σ_d, see Fig. 4).

As indicated in Fig. 4a, the "plaquette spin-nematic" state covers a wide range $0.40 \lesssim J'/J \lesssim 0.66$. It has been shown in Ref. [29] that the $\frac{1}{8}$ plateau at slightly higher magnetic field values and $J'/J = 0.63$ is induced by crystallization of $S^z_{\rm tot} = 2$ bound states. A closer scrutiny of the "pinwheel" structure of these bound states shows that they locally preserve rotational symmetries, while breaking reflection symmetries [29], i.e., they are the same two-triplon bound states that we are finding in the dilute limit.

Moving away from the plaquette spin-nematic phase towards the $J'/J \ll 1$ limit, it is already known from an early perturbative calculation that $S_{\rm tot} = 2$ two-triplon bound states with $\boldsymbol{k} = (\pi,\pi)$ give the highest susceptibility [19]. This is confirmed by our variational method (see Fig. 3d). Since the two-triplon bound state has momentum $\boldsymbol{k} = (\pi,\pi)$, the corresponding ordered state also breaks translational symmetry. As shown in Fig. 4e, $\langle S_i^+ S_j^+ \rangle$ changes sign going from one unitcell to its nearest neighbors. Similar to the case of the stripe ordering (which also comes from condensation of IREP E states), there are two choices for aligning the bonds. We note that the breaking of the C_4 lattice rotational symmetry leads to a modulation of $\langle S_i^z \rangle$ that can be detected with NS experiments.

It is worth mentioning that the two spin-nematic states found in this Letter are different from nematic phases discussed in various other contexts [78–81]. The so-called "Ising-nematic" ordering corresponds to (discrete) lattice rotation symmetry breaking. In contrast, "spin-nematic" ordering corresponds to broken spin rotational symmetry. The spin-nematic orderings discussed in this Letter *break both the point group symmetry and spin rotation symmetry* [82]. In other words, they are *simultaneously* Ising-nematic and spin-nematic.

The frustrated nature of the SSM makes the calculation of dynamical response a difficult task. Up to date, the only calculation including multi-triplon contributions is the perturbative CUTs, which breaks down for $J'/J \gtrsim 0.63$ [83]. The variational Hilbert space generated by our method thus provides a more reliable access to dynamical responses via the continued fraction method [84].

Near the phase boundaries, we expect the susceptibilities of corresponding order parameters to diverge at $\omega = 0$. Magnetic

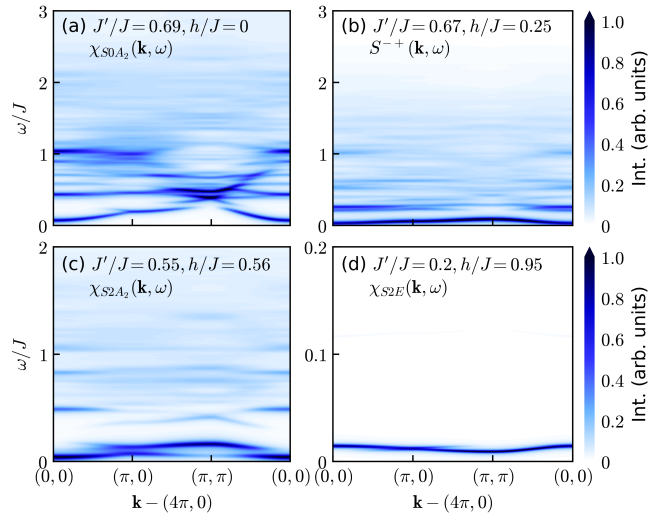

Figure 5. $T = 0$ DSFs calculated near the phase boundaries at $M = 8$. (a-c) Lorentzian broadening factor $\eta = 0.02J$ is used. (d) Lorentzian broadening factor $\eta = 0.001J$ is used.

orderings, such as the stripe phase, are detected by computing the dynamic structure factor (DSF) [70, 85]:

$$S^{-+}(\boldsymbol{k}, \omega) = 2\pi \sum_{\nu} |\langle \nu | S^{+}_{\boldsymbol{k}} | 0 \rangle|^2 \delta(\omega + E_0 - E_\nu), \qquad (3)$$

which is measured with inelastic NS. As shown Fig. 5b, the lowest peak of $S^{-+}(\boldsymbol{k}, \omega)$ approaches $\omega = 0$ near the phase boundary indicating condensation of an $S_{\text{tot}} = 1$ state.

The divergent susceptibilities of the other phases are revealed by computing two-point dynamical correlation functions of the corresponding order parameters. These order parameters are the operators that create a state that has finite overlap with the one that is condensing. For $J'/J \ll 1$, the lowest energy $S_{\text{tot}} = 2$ eigenstates are known to be a linear combination of triplons located on nearest (and next-nearest) neighbors [19, 70]. Denoting the order parameter as $A^{S2E}_{\boldsymbol{k}}$, the corresponding susceptibility is

$$\chi_{S2E}(\boldsymbol{k}, \omega) = 2\pi \sum_{\nu} |\langle \nu | A^{S2E}_{\boldsymbol{k}} | 0 \rangle|^2 \delta(\omega + E_0 - E_\nu). \qquad (4)$$

Similarly, using the approximate wavefunctions of the $S_{\text{tot}} = 2$ A_2 state and the $S_{\text{tot}} = 0$ A_2 state [70], we can also construct the order parameters and compute the corresponding susceptibilities $\chi_{S2A_2}(\boldsymbol{k}, \omega)$ and $\chi_{S0A_2}(\boldsymbol{k}, \omega)$. Figure 5 shows the nearly divergent susceptibilities in each channel by picking appropriate Hamiltonian parameters near the phase boundaries.

While the tendency toward stripe ordering can be detected with inelastic NS, the experimental detection of the other phases is nontrivial. Lattice distortions induced by the order parameter through magnetostriction can provide indirect evidence if they are large enough to be detected [75–77]. Experimental knobs, such as pressure, doping and magnetic field, can drive the material into different instabilities [39, 47]. Thus, the method presented in this Letter provides valuable insight for revealing the nature of the new phases in such experiments. However, the model relevant to $SrCu_2(BO_3)_2$ also includes DM interactions that modify the single triplon dispersion and can potentially change the phase diagram reported here. In addition, DM interactions reduce the spin rotational symmetry of the model, implying that they can change the nature of the order parameters.

We emphasize that the applicability of the method is not restricted to the SSM considered here. The same method can be used to detect the instabilities of other quantum paramagnets [76]. Especially, it is very difficult to enumerate all the possible instabilities for highly frustrated systems. The low energy spectrum produced by our method provides a valuable educated guess for biased numerical approaches. Given that the method works in the thermodynamic limit, it can also detect incommensurate instabilities, which cannot be handled by most numerical methods.

In summary, we have used an unbiased variational method to study the excitation spectrum of the SSM in the dimer phase. Several instabilities are found next to the dimer phase corresponding to condensations of single-triplon or two-triplon bound states. Two of the instabilities (antiferro spin-nematic and plaquette) are known from previous studies and the others (plaquette spin-nematic and stripe) are newly discovered in this Letter. The same method can be used to compute relevant dynamical correlation functions.

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[69] This means that the basis generates an invariant subspace of the group. In addition, a fully symmetric initial basis leads to better convergence (as a function of M) in comparison with other choices Ref. [65].
[70] See Supplemental Material for the C_{4v} character table, choice of initial basis, construction of the order parameters, dominant instability at zero field, convergence of dynamic structure factors, and the description of a competing chiral state, which includes Ref. [91].
[71] The computational cost is exponential in the *linear* size of the quasi-particle. In the present case, the dimension of the truncated Hilbert space roughly scales as $\mathcal{D} \propto 6^M \propto 6^l$. For the $S^z_{\rm tot} = 0$ sector, the dimension of the variational space is $\mathcal{D} = 74$ for $M = 1$, $\mathcal{D} = 396$ for $M = 2$, ..., $\mathcal{D} = 17730330$ for $M = 8$.
[72] In principle, one can switch the order of "diagonalization" and "point group symmetry analysis", to reach larger M and get better convergence. Similar tricks can be found in Ref. [92, 93].
[73] The $S = 1$ instability found in Ref. [7] is checked by our method both in $S^z_{\rm tot} = 0$ and $S^z_{\rm tot} = 1$ sectors. The energy difference between the two sectors is negligible.
[74] A typical example is the case of attractive interaction between the modes (particles) that become soft. Because of the attraction (negative quartic term in a Ginzburg-Landau expansion) the particle density changes discontinuously (first order transition) before the single-particle excitation becomes gapless.
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