Triangular Lattice Quantum Dimer Model Redux: Static and Dynamic Properties

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Quantum dimer model (QDM) is the prototypical correlated system with local constraints, whose generic solution at different lattice geometry and parameter regimes is still missing due to the lack of controlled methodology. Here we obtain, via the newly developed sweeping cluster quantum Monte Carlo algorithm, the excitation spectra in different phases of the triangular lattice QDM. Our results reveal the single vison excitations inside the $Z_2$ quantum spin liquid (QSL) and its condensation towards the $\sqrt{12}\times\sqrt{12}$ valence bond solid (VBS), and demonstrate the translational symmetry fractionalization and emergent $O(4)$ symmetry at the QSL-VBS transition. We find that the single vison excitations, whose convolution qualitatively reproduces the dimer spectra, are not free but subject to interaction effects throughout the transition. The nature of the $\sqrt{12}\times\sqrt{12}$ VBS with its $O(4)$ order parameters are unearthed in full scope. Our approach opens the avenue for complete solution of the static and dynamic properties of QDMs.

Introduction.- Fractionalized anyon excitations are one of the most important features of topologically ordered phases, a class of phases beyond the Landau paradigm of classifying phases with symmetry breaking [1]. The fractionalized nature of these anyon excitations renders that they cannot be created or annihilated individually by physical probes. This phenomenon is both a bless and a curse: it reflects the topological nature of the excitations and the phase, but also obscures any direct detection of single anyon excitations. Instead, they can only be observed indirectly from a multi-particle continuum of spectral functions. For example, a continuum in inelastic neutron scattering spectrum is often used as a signature to detect quantum spin liquids with fractionalized spin excitations, which is considered as a two-spinon continuum [2,3]. Consequently, understanding the relation between physical spectra and underlying single-anyon excitation is an essential question in the study of topologically ordered phases.

In the theoretical study of topologically ordered phases including the quantum spin liquid (QSL), one usually relies on approximate tools to model the fractionalized excitations because they cannot be directly accessed in experiments and numerical simulations. In simple mean-field theories of QSL, as a physical probe excites a pair of fractionalized excitations, the corresponding spectrum is given by the convolution of spectra of the underlying anyons. However, in realistic systems, this simple relation is modified by interactions between anyons, it is therefore important to know how much change has happened due to the interaction effect.

Quantum dimer models (QDM) [4,5] provide a unique playground for studying the relation between single-anyon and two-anyon spectra in QSLs. Originally proposed to model the resonant valence bond state in high-$T_c$ superconductors [6] and frustrated magnets, it realizes a gapped $Z_2$ quantum spin liquid at the exactly-solvable Rokhsar-Kivelson (RK) point [5] if put on a nonbipartite lattice such as the triangle and the kagome [7,8]. Comparing to other models of QSLs, the QDMs are unique as the spinful excitations are absent in the Hilbert space due to the one-dimer-per-site constraint. In particular, this means that the spinon excitations in the $Z_2$ spin liquid are absent, leaving the visons as the only low-energy anyon excitations. As a result, the spectrum of vison excitations can be directly measured in numerical simulations. This feature of QDM allows one to compare the spectra of both the fractionalized single-vison excitations and the physical dimer-dimer correlations, which involves a pair of visons. Although the ground state of QDM is exactly known at the RK point, the excited states are not exactly solvable due to interactions among the visons.

Furthermore, away from the RK point, the QDM on the triangular lattice can be tuned into a $\sqrt{12}\times\sqrt{12}$ valence bond solid (VBS) phase [7,8]. The phase transition is conjectured to be continuous, and belonging to the $O(4)$ universality, driven by the condensation of visons [9,10]. Therefore, the QDM near this transition is an ideal system to study the spectral properties of anyon condensation, if there exist controlled theoretical and numerical methods.

Recently, a new quantum Monte Carlo scheme, the sweeping cluster method, is invented by one of us [13]. The method is able to keep track of the strict local constraint of dimer covering and at the same time perform Markov chain Monte Carlo in the space-time path integral such that both static and dynamic properties of the QDM can be obtained, only subject to finite system sizes. The method has been applied to the square lattice QDM and a mixed phase separating columnar phase at strong
dimer attraction and staggered phase at strong dimer repulsion are found [14]. In this work, we further develop the method and employ it to the static and dynamic properties of triangular lattice QDM.

The problem has a long and interesting history. From the work of Moessner-Sondhi [2], one knows that from the mapping to frustrated Ising model on honeycomb lattice, the problem is in principle solvable via Monte Carlo simulations on the frustrated Ising model, and a $\sqrt{12} \times \sqrt{12}$ VBS and a $Z_2$ QSL are suggested. Then in a series of works with zero temperature Green’s function Monte Carlo [9–12], the transition from the QSL to VBS, with the notion that the gap of topological vison excitations is closed at the transition is revealed, although the numerical method therein could only work close to the RK point and zero temperature. Later, the dynamical dimer correlations at the RK point is presented in Ref. [15], taking the advantage that at the RK point, the quantum mechanics in imaginary time among the equally weighted dimer coverings is equivalent to a classical stochastic process [16]. Despite of these important progresses, the complete spectra of both dimer and vison excitations, not only the gap but also the spectral weight, and the complete understanding of the transition from $Z_2$ QSL to the $\sqrt{12} \times \sqrt{12}$ VBS in the modern language of topological order and symmetry fractionalization, and the nature of the complex $O(4)$ order parameter of the VBS, are not revealed. Here we try to answer these questions with unbiased QMC and symmetry analysis.

Model and Measurements.- We study the QDM on triangular lattice with the Hamiltonian,

$$H = -t \sum_r \left( |\langle \uparrow \uparrow \rangle | \langle \downarrow \downarrow | + h.c. \right) + V \sum_r \left( |\langle \uparrow \downarrow \rangle | \langle \uparrow \downarrow | + |\langle \downarrow \uparrow \rangle | \langle \downarrow \uparrow | \right)$$  

(1)

where the sum runs over all plaquettes (rhombi) including the three possible orientations. The kinetic term, controlled by $t$, flips the two dimers on every flippable plaquette, i.e., on every plaquette with two parallel dimers, while the potential term controlled by the interaction $V$ describes a repulsion ($V > 0$) or an attraction ($V < 0$) between nearest-neighbor dimers. We set $t = 1$ as the energy unit throughout this paper.

Before the sweeping cluster QMC [13–14], one commonly employs the projector approaches to study QDMs, which includes the Green’s function [9–12] and diffusion Monte Carlo schemes [17–18]. These projector methods obey the geometric constraints, but are not effective away from RK point [19] and only work at $T = 0$. Also, there exists no cluster update for the projector methods to reduce the computational complexity. On the contrary, the sweeping cluster algorithm is based on world-line Monte Carlo scheme and it contains cluster update scheme for constrained systems and works at all temperatures.

The key idea of sweeping cluster algorithm is to sweep and update layer by layer along the imaginary time direction, so that the local constraints are recorded by update-lines. It is an extension of the directed-loop algorithm [20] employed to compute the D dimension classic dimer model [21] to the quantum dimension of imaginary time at $(D+1)$. As for the spectral functions, we employ the stochastic analytic continuation (SAC) method [22–27] to obtain the real frequency excitation spectra from their QMC imaginary correlation functions. Details of the implementation of sweeping cluster QMC and SAC can be found in the following literatures [13–14] [22–29].

We compute three different dynamical correlation functions in QMC. The first one is the dimer correlation. The dimer operator $D_i = 1$ when there is a dimer on the link $i$, and $0$ otherwise. The dimer correlation function is then defined as $C_d(r_{i,j}, \tau) = \langle D_i(\tau)D_j(0) \rangle = \langle D_i \rangle^2$, average over the lattice, and $C_d(q, \tau)$ through the Fourier transformation, then the excitation spectrum $C_d(q, \omega)$ via SAC.

The second one is the vison correlation. Visons ($V_i$) live in the centre of triangle plaquettes and they must arise in pairs, as shown in the right inset of Fig. 1(c). The correlation func-
FIG. 2. Spectra of dimer (a) (b) (c), vison pair (d) (e) (f) and vison (g) (h) (i) correlation functions across the VBS-QSL transition. For (a) (d) (g), (b) (e) (h) and (c) (f) (i), $V = 0.8, 0.9, 1$, respectively. The results are obtained from $L = 12$ and $\beta = 200$ systems.

The last one is the vison pair correlation function. We denote the vison pair operator as $D_{ij}^{vp} = V_{i1}V_{j2}d_i d_j$, i.e. if two visons are close to each other, then there is only one link between them and the dimer operator can be represented as these two vison operators, with $i_1$ and $i_2$ the triangle plaquettes closest to the link $i$. $d_i = \pm 1$ when there is no / one dimer on link $i$ of the reference configuration. Then the correlation function is $C_d^{vp}(r_i, r_j, \tau) = \langle D_{ij}^{vp}(0)D_{ij}^{vp}(\tau) \rangle - \langle D_{ij}^{vp}(0) \rangle^2 = \langle V_{i1}(0)V_{j2}(0)d_i d_j V_{i1}(\tau)V_{j2}(\tau) d_i d_j \rangle - \langle V_{i1}(0)V_{j2}(0) \rangle^2$. Using Wick’s theorem, we arrive at

$$C_d^{vp}(r_i, r_j, \tau) = \langle V_{i1}(0)V_{j1}(\tau) \rangle \langle V_{i2}(0)V_{j2}(\tau) \rangle d_i d_j + \langle V_{i1}(0)V_{j1}(\tau) \rangle \langle V_{i2}(0)V_{j2}(\tau) \rangle d_i d_j$$

where the difference will reveal the interaction effects among the visons in different regions of the phase diagram.

**Spectra of dimer, vison and vison pair.** In the $Z_2$ QSL phase, the visons are the emergent and fractionalized elementary excitation with no spin and charge quantum numbers. As discussed in the introduction, this is an unique advantage of the QDM that single vison spectrum can be measured unambiguously, as usually the vison excitations have to be constructed in mean-field as built-in without knowing the unbiased physics [30], or measured indirectly in lattice models of frustrated magnets [25, 31-34].

We therefore measure the correlation functions of $C_d(q, \omega)$, $C_v(q, \tau)$ and $C_d^{vp}(q, \omega)$ in QMC and then using SAC [23, 25] to generate the real frequency spectra $C_d(q, \omega)$, $C_v(q, \omega)$ and $C_d^{vp}(q, \omega)$. These results are presented in Fig. 2. Inside the $Z_2$ QSL phase with $V = 1$, all the spectra are gapped. The vison spectra acquire the smallest gap at the order of $\omega \sim 0.1$. 

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The correlation function is defined as $V_i V_j = (-1)^{N_{R_{ij}}}$ where $N_{R_{ij}}$ is the number of dimers along the path $P_{ij}$, we chose between plaquettes $i$ and $j$ as shown in Fig. 1 (c). It is clear that the value of $V_i V_j$ is path dependent. In order to eliminate this dependence, one can choose a reference configuration, and follow the same path $P_{ij}$ again to obtain another $N_{R_{ij}}$ and then the new observable $N_{R_{ij}} = N_{R_{ij}} - N_{R_{ij}}$ is path independent. Then we redefine $C_r(r_i, r_j, \tau) = \langle V_r(\tau)V_r(0) \rangle = \langle V_r(\tau)V_r(0)V_r(0)V_r(\tau) \rangle = \langle (-1)^{N_{R_{eff}} + N_{R_{eff}}} \rangle$ where $N_{R_{eff}}$ means the number of off-diagonal Hamiltonian operators, i.e. the $t$-term in Eq. [1], between $V_i(\tau)$ and $V_i(0)$. We choose the reference configuration as the columnar VBS shown in Fig. 1 (a), which doubles the unit cell and the corresponding BZ under this reference (gauge choice) is the dashed rectangle with high symmetry points A, B and C in Fig. 1 (b).
at the $B$ point inside its BZ. And the dimer and vison pair correlations are also gapped with their minimal at $M$ point. It is interesting to notice that the vison pair spectral gap at $M$ point is higher than the dimer gap at the same point, suggesting that actually visons have a binding energy in forming the dimer correlation and consequently their interaction effect is attractive and give rise to a bound state with lower energy than the naive convolution.

FIG. 3. (a), (b) are the projection of the four-dimensional order parameter $(w, x, y, z)$ at the QSL-VBS critical point with $V = 0.85$ on the two-dimensional $(w, x)$ plane (a) and on the $(y, z)$ plane (b). (Inset) The density distribution per unit sphere area of the O(4) order parameter modulus at $V = 0.85$, such radial dependence reveals that the order parameter indeed form a O(4) sphere. (c) and (d) are the projection of the four-dimensional order parameter deep in the $\sqrt{12} \times \sqrt{12}$ VBS phase at $V = 0$. The histogram is on the $(w, x)$ plane (c) and the $(y, z)$ plane (d). The data in (a), (b), (c) and (d) are obtained from system size $L = \beta = 36$.

As $V$ is reduced from 1 to 0.9 and 0.8, a QSL-VBS transition is expected at $V_c \sim 0.85$ \cite{9,12}, and previous works from the gap measurements and field analytical analysis \cite{7} have proposed emergent O(4) symmetry at the transition. But how the entire spectra change across the transition has not been shown due to the lack of access to finite temperature fluctuation effects. With our QMC+SAC scheme, we observe that the vison gap closes at the $B$ point and the dimer and vison pair gap close at $X$ and $M$ points of the BZ (subject to finite size effect of the QMC simulation), as shown in Fig. 2 for $V = 0.8$ and 0.9. The minimal at $\Gamma$ and $K$ of the vison pair spectra come from the allowed momentum convolution of single vison spectra which has minimal at $B$. Such gap closing process is a manifestation of the symmetry fractionalization mechanism of anyon condensation in $Z_2$ topological order \cite{25,35,36}. That is, since here the $Z_2$ gauge field is odd in nature (see the discussion in Supplemental Material (SM) \cite{37}), the visons carry $\pi$-flux throughout the lattice. This means that as the QSL-VBS critical point is approached, the vison gap will close and the entire vison spectral weight will condensed at a finite momentum point. In a similar manner, the dimer spectra, generated from the vison bound states, will also close at a finite momentum point. This is different from the usual Bose condensation from disorder symmetric state to ordered symmetry-breaking state, where the condensation of the low-lying bosons usually close gap at the $\Gamma$ point. Since in our case the disordered state has intrinsic topological order with elementary excitations (visons) carrying finite moment (i.e., $\pi$-flux), the condensation gap manifests finite momentum closing. Also, one sees that at $V = 0.9$ and 0.8, there are more higher energy spectral weights in dimer, vison pair and vison spectra, coming from the enhanced quantum critical fluctuations of the QSL-VBS transition.

Emergent O(4) symmetry and order parameter of VBS. Next we discuss the nature of the QSL-VBS transition and the symmetry breaking pattern of the $\sqrt{12} \times \sqrt{12}$ phase. As explained in the SM \cite{37}, it is expected theoretically \cite{8} that this transition is driven by the condensation of visons, which is described by a four-component order parameter $\{\phi_i\}, i = 0, 1, 2, 3$ constructed from the Fourier transformation vison configuration at momenta $B$, i.e., $\pm (\frac{\pi}{6}, \frac{\pi}{6})$ and $\pm (\frac{\pi}{6}, -\frac{\pi}{6})$ in Fig. 1(b). The order parameter transforms as a 4D representation under the lattice wall-paper-group symmetries, and the matrix form of group actions are summarized in the SM \cite{37}.

In order to numerically confirm that the order parameter $\phi_i$ indeed captures the QSL-VBS transition, we perform a principal component analysis (PCA) on the vison correlation function $C_i$ to extract the condensing mode near the transition. PCA diagonalizes the $4 \times 4$ matrix of the momentum-space vison correlation function at the $B$ point, and identifies the eigenvectors with the largest eigenvalues corresponds to the modes represented by the order parameter $\phi_i$. We list the ratio of the first largest eigenvalue over the second at $V = 0.5, 0.6, 0.7, 0.8, 0.9, 1$ in Table. 1. Since the largest eigenvalue always dominate, it shows that the principal component of the VBS structure is indeed the expected $\sqrt{12} \times \sqrt{12}$ order. The theoretical analysis further predicts that, at the QSL-VBS critical point, the transition point acquires an emergent O(4) symmetry, as O(4)-symmetry-breaking terms are all irrelevant at the critical point. In other words, the order parameter lives homogeneously on a four-dimensional sphere \cite{7}.

| $V$  | 0.5  | 0.6  | 0.7  | 0.8  | 0.9  | 1   |
|------|------|------|------|------|------|-----|
| $L_1/L_2$ | 85.03 | 82.05 | 76.18 | 68.48 | 51.36 | 29.71 |

TABLE I. Principal Component Analysis. $L_1/L_2$ means the first largest eigenvalue over the second one of the momentum-space vison correlation function matrix at $B$ point. All data are obtained at a $12 \times 12$ lattice with $\beta = 200$.

To reveal such emergent O(4) symmetry at the QSL-VBS critical point and its breaking inside the $\sqrt{12} \times \sqrt{12}$ VBS phase. We prepare the order parameter histogram in Fig. 3.
By reorganizing the order parameters into $\phi_0 = \phi_2^* = w + ix$ and $\phi_1 = \phi_3^* = y + iz$. We can draw two-dimensional projected histogram $(w, x)$ and $(y, z)$ of the 4D order parameter near the phase transition point at $V = 0.85$ and deep inside the VBS phase at $V = 0$. Fig. 3(a) and (b) are the two independent projections of the 4D $(w, x, y, z)$ space and clearly an emergent $O(4)$ symmetry is present. The Inset shows the modulus distribution of the 4D sphere which means the order indeed lives homogeneously on a four-dimensional sphere. Fig. 3(c) and (d) are the same analysis inside the VBS phase, and here clearly distinctive points in the two projected phase are present, which are in full consistency with the analysis in SM [37], i.e. the $\sqrt{12} \times \sqrt{12}$ VBS breaks the $O(4)$ symmetry.

Conclusion.- Via the newly developed sweeping cluster QMC algorithm, supplemented with SAC scheme to obtain the real-frequency data and symmetry analysis of the VBS order parameter, we reveal the excitation spectra in different phases of the triangular lattice QDM, in particular, the single vison excitations inside the $Z_2$ QSL and its condensation towards the $\sqrt{12} \times \sqrt{12}$ VBS with the translational symmetry fractionalization. We found the vison pair spectrum is different from the dimer spectrum due to the vison interaction effect, and we also unearth the emergent $O(4)$ symmetry at the QSL-VBS transition and the nature of the $\sqrt{12} \times \sqrt{12}$ VBS with its $O(4)$ order parameter and symmetry breaking. Our approach opens the avenue for generic solution of the static and dynamic properties of QDMs.

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[37] Derivations of the vison operators, the $O(4)$ order parameter of the VBS phase and its symmetry transformation.
SUPPLEMENTAL MATERIAL

Triangular Lattice Quantum Dimer Model Redux: Static and Dynamic Properties

In this supplemental material, we analyze the symmetry of the vison order parameter, associated with the phase transition between the $\mathbb{Z}_2$ QSL and the $\sqrt{12} \times \sqrt{12}$ VBS phases in the QDM on triangular lattices. Intuitively, such a phase transition is driven by the condensation of visons, which belong to a type of fractional excitations in the $\mathbb{Z}_2$ QSL. The correlation function of visons can be described using the vison-string operator, defined on a string driven by the condensation of visons, which belong to a type of fractional excitations in the $\mathbb{Z}_2$ QSL. The correlation function of visons can be described using the vison-string operator, defined on a string $L : I \rightarrow J$ connecting two sites of the dual lattice:

$$ V_L = \prod_{(ij) \in L} (-1)^{n_{ij}}, \quad (S1) $$

where the product goes over each bond $\langle ij \rangle$ cut by the string $L$ on the dual lattice, as shown in Fig. S1(a).

**VISON OPERATORS**

To analyze the vison condensation, it is convenient to map the quantum dimer model to a fully-frustrated transverse-field Ising model (FFTFIM) on the dual lattice, which is a honeycomb lattice [7]. The mapping is illustrated in Fig. S1(b): Two spins on nearest-neighbor sites are frustrated (unfrustrated) if there is a dimer (no dimer) on the bond separating them, respectively. In other words, the spins and dimer occupation satisfies the following relation,

$$ J_{IJ} s_i^x s_j^x = (-1)^{n_{ij}}. \quad (S2) $$

Here, the bond $\langle ij \rangle$ separates the two dual-lattice sites $I$ and $J$. $J_{IJ}$ is a fully-frustrated Ising coupling: there is exactly one antiferromagnetic coupling $J_{IJ} = -1$ on each hexagon of the dual honeycomb lattice. The pattern of $J_{IJ} = \pm 1$ is a choice of gauge. Without losing generality, we follow Ref. [7] but choose another gauge shown in Fig. S1(c). This mapping is neither surjective nor injective. On one hand, it maps each dimer configuration to two spin configurations that are exactly opposite to each other, because the two spin configurations $(s_i^x, s_j^x)$ and $(−s_i^x, −s_j^x)$ give the same $n_{ij}$ in Eq. (S2). On the other hand, dimer configurations satisfying the one-dimer-per-site rule only maps to a subset of spin configurations which minimize the energy of the fully-frustrated Ising model (FFIM). In this way, the original quantum dimer model can be mapped to the following FFTFIM,

$$ H = - \sum_{\langle ij \rangle} J_{IJ} s_i^x s_j^x - \Gamma \sum_I s_i^z, \quad (S3) $$

in the small-$\Gamma$ limit, $\Gamma \ll |J_{IJ}|$. On the level of Hamiltonians, the mapping between the QDM and the FFTFIM is only approximate and only valid at one parameter point. However, on the level of quantum states, the mapping is always valid between dimer configurations and spin configurations in the low-energy sector. Therefore, the map can be used to construct order parameters and analyze the phase transition of vison condensation at different parameters across the phase transition.

Motivated by the mapping in Eq. (S2), we construct vison creation/annihilation operators in the QDM from the string operator in Eq. (S1). The $\mathbb{Z}_2$ spin liquid phase maps to the paramagnetic phase in the FFTFIM, which is realized in the strong-$\Gamma$ limit, $\Gamma \gg |J_{IJ}|$. The ground state in this limit has all spins pointing to the $x$ direction, $s_i^z = +1$. A vison excitation is a flipped spin, $s_i^z = -1$. Hence, the vison creation/annihilation operator is $s_i^z$. Note that the creation and annihilation operator is the same, because visons obey a $\mathbb{Z}_2$ fusion rule and they are their own antiparticle. The relation in Eq. (S2) then implies that the dimer-parity operator $(-1)^{n_{ij}}$ is a product of two vison creation/annihilation operators: it creates/annihilates two visons on the two neighboring dual sites, or moves a vison between them. Using this relation repeatedly, we can map the two-point operator $V_{IJ} = s_i^z s_j^z$ to a string of dimer-parity operators. We first choose an arbitrary string $L : I \rightarrow J$ connecting the two dual sites $I$ and $J$. Following $L$, we rewrite the correlation function as a string operator,

$$ V_{IJ} = \prod_{\langle ij \rangle \in L} s_i^z s_j^z. $$

Using the relation Eq. (S2), we rewrite this using the dimer-parity operators on the bonds $\langle ij \rangle$ the string $L$ cuts through,

$$ V_{IJ} = \prod_{\langle ij \rangle \in L} (-1)^{n_{ij} + n_{ij}^\prime}, \quad (S4) $$
FIG. S1. (a) Vison-string operator and cut links. (b) Map the quantum dimer model to a fully-frustrated transverse-field Ising model (FFTFIM) on the dual lattice. (c) The reference chosen for vison. 0, 1, 2, 3 four sublattice of vison. The x and y axis of triangular lattice.

where \( n'_{ij} = 1 \) or \( 0 \) is determined by \((-1)^{n'_{ij}} = J_{IJ}\). Here, \( n'_{ij} \) can be viewed as a reference dimer configuration, determined by the gauge choice of \( J_{IJ} \), as shown in Fig. S1(c). Comparing to the vison string operator in Eq. (S1), the two operators give the same physical result, because they only differ by a static sign independent of the physical dimer configuration. However, the operator in Eq. (S4) has the advantage that it has no dependence on the choice of the path \( L \), whereas the operator Eq. (S1) has a weak dependence on \( L \): the operators defined by two paths \( L \) and \( L' \) encloses even (odd) number of sites, respectively. The independence of the choice of paths makes the operator in Eq. (S4) convenient for studying the position-dependence and the Fourier transform of \( V_{IJ} \). Next, we introduce a vison creation/annihilation operator in the QDM. In the FFTFIM, this operator can be obtained by multiplying the two-point operator \( s_z^I s_z^J \) with \( s_z^J \), where the site \( J = 0 \) is an arbitrary reference point. Since the mapping from dimer to spin configurations is two-to-one, one can choose \( s_z^0 = \pm 1 \) arbitrarily and obtain the following vison creation/annihilation operator in the QDM,

\[
\hat{V}_I = \hat{V}_{10}\eta,
\]

where \( \hat{V}_{10} \) is a string operator defined in Eq. (S4) connecting the dual site \( I \) to the reference dual site \( J = 0 \), and \( \eta = s_z^J = \pm 1 \) is an arbitrarily chosen sign. Using this vison operator, it is easy to verify that the vison string operator can be expressed as

\[
\hat{V}_{IJ} = \hat{V}_I \hat{V}_J,
\]

as expected. The choice of \( \eta = \pm 1 \) does not affect the calculation of any correlation functions, but one need to count both choices to obtain a histogram of the hidden order parameter, as we will discuss later.

ORDER PARAMETER

We now construct the order parameter of the vison-condensation phase transition. Following Ref. [7], we begin by studying the dispersion relation of the vison excitations in the \( \Gamma \gg J \) limit. In the limit of \( J = 0 \), the vison excitation is a single flipped spin \( s_z^I = -1 \), with energy gap \( \Gamma \). In the next order, the Ising coupling \( -J_{IJ} \) can be viewed as a hopping Hamiltonian for the visons, and gives them a dispersion. After Fourier transformation, it can be written in the following matrix form,

\[
H_J = \sum_{k,a} s_q^a H(q) s_q^a,
\]

where \( s_q^a \), the Fourier transform of \( s_I^a \), is a four-component vector: \( s_q^a = \left( s_{q0}^a, s_{q1}^a, s_{q2}^a, s_{q3}^a \right)^T \). Here, the four components \( a = 0, \ldots, 4 \) labels the four sublattices, as shown in Fig. S1(c). (The dual honeycomb lattice originally has two sublattices, but the pattern of fully frustrated \( J_{IJ} \) further doubles the unit cell.) If we label dual-lattice sites \( I \) by the combination of \( \bar{I} \) and \( a \), which labels the unit cell and the sublattice, respectively, the Fourier transform can be expressed as

\[
s_{qa}^I = \sum_{\bar{I}} s_{\bar{I}a}\ e^{-i\bar{r}_I \cdot q}.
\]
Here, the Fourier transform is performed using the positions of the unit cell, \( r_I \), instead of the position of the actual site. In this convention, the matrix \( H(q) \) has the following form,

\[
H = \sum_{q} E_{q\alpha} \tilde{s}_{q\alpha} \tilde{s}_{q\alpha}.
\]

(S9)

where \( \tilde{s}_{q\alpha} \) is related to \( s^z_{q\alpha} \) by the unitary transformation

\[
s^z_{q\alpha} = \sum_{\alpha} u_{q\alpha,\alpha} \tilde{s}_{q\alpha}.
\]

(S11)

Therefore, the operators \( \tilde{s}_{q\alpha} \) represents the vison modes in this model. In particular, we focus on the lowest band, which we denote by \( \alpha = 0 \). The dispersion \( E_{q0} \) has minima at four momenta: \( Q_{0,2} = \pm \left( \frac{\pi}{\Gamma}, \frac{\pi}{6} \right) \) and \( Q_{1,3} = \pm \left( -\frac{\pi}{\Gamma}, \frac{\pi}{6} \right) \), the \( B \) point in the BZ as shown in Fig. 1(b) of the main text, where the minimum energy is \( \Gamma = \frac{\sqrt{6}}{4} J \). The eigenvectors at these momenta are

\[
u_{Q_{0,0}} = u^*_{Q_{0,0}} = \left( \frac{1}{\sqrt{\Gamma}} e^{i\frac{\pi}{\Gamma}}, 1, e^{-i\frac{\pi}{6}}, e^{i\frac{\pi}{6}} \right)^T,
\]

\[
u_{Q_{0,1}} = u^*_{Q_{0,1}} = \left( e^{-i\frac{\pi}{6}}, 1, e^{-i\frac{\pi}{6}}, e^{i\frac{\pi}{6}} \right)^T.
\]

(S12)

At the vison-condensation transition, the vison condenses at the four modes above. Therefore, the order parameter of this phase transition is the expectation values of the corresponding \( \tilde{s}_{Q_{0,0}} \) operators. Furthermore, in order to construct a Ginzburg-Landau theory with a position-dependent order-parameter field, we also consider momenta close to but not exactly at \( Q_{1} \). Hence, we introduce the following fields,

\[
\phi_i(k) = \tilde{s}_{q_{i,0} + k,0}.
\]

(S13)

Here, \( k \ll 1 \) is a small momentum. Fourier-transforming \( \phi_i(k) \) yields the real-space order parameter \( \phi(\mathbf{r}) \). Here, \( \mathbf{r} \) still labels the location of the unit cell, as \( k \) is a momentum in the Brillouin Zone. Hence, we denote the real-space order parameter as \( \phi_i(\bar{I}) \), where \( \bar{I} \) labels the unit cell.

\[
\phi_i(\bar{I}) = \sum_k \tilde{s}_{q_{i,0} + k,0} e^{i k \cdot r_I} = \sum_{\alpha} u^*_{Q_{0,0},\alpha} e^{-i Q_{0,0} \cdot \mathbf{r}_I} s^z_{\alpha}.
\]

(S14)

Plugging in Eq. (S12), we get

\[
\phi_0(\bar{I}) = \phi_2(\bar{I})^* = \left( \frac{1}{\Gamma} e^{i\frac{\pi}{\Gamma}} v_{10} + \frac{1}{\Gamma} e^{-i\frac{\pi}{\Gamma}} v_{11} + e^{-i\frac{\pi}{6}} v_{12} + e^{i\frac{\pi}{6}} v_{13} \right) e^{-i\frac{\pi}{6} 2\bar{X}_{10} - i\frac{\pi}{6} 2\bar{X}_{11}}.
\]

\[
\phi_1(\bar{I}) = \phi_3(\bar{I})^* = \left( e^{-i\frac{\pi}{6}} v_{10} + e^{-i\frac{\pi}{6}} v_{11} + \frac{1}{\Gamma} e^{-i\frac{\pi}{6}} v_{12} + \frac{1}{\Gamma} v_{13} \right) e^{i\frac{\pi}{6} 2\bar{X}_{10} - i\frac{\pi}{6} 2\bar{X}_{11}}.
\]

(S15)

Replace the spin operators \( s^z_{\alpha} \) by the vison operators in Eq. (S5), we can compute the order parameters \( \phi_i \) in QDM as

\[
\phi_0(\bar{I}) = \phi_2(\bar{I})^* = \left( \frac{1}{\Gamma} e^{i\frac{\pi}{\Gamma}} v_{10} + \frac{1}{\Gamma} e^{-i\frac{\pi}{\Gamma}} v_{11} + e^{-i\frac{\pi}{6}} v_{12} + v_{13} \right) e^{-i\frac{\pi}{6} 2\bar{X}_{10} - i\frac{\pi}{6} 2\bar{X}_{11}}.
\]

\[
\phi_1(\bar{I}) = \phi_3(\bar{I})^* = \left( e^{-i\frac{\pi}{6}} v_{10} + e^{-i\frac{\pi}{6}} v_{11} + \frac{1}{\Gamma} e^{-i\frac{\pi}{6}} v_{12} + \frac{1}{\Gamma} v_{13} \right) e^{i\frac{\pi}{6} 2\bar{X}_{10} - i\frac{\pi}{6} 2\bar{X}_{11}}.
\]

(S16)

Again, we emphasize that, to verify the emergent O(4) symmetry in \( \phi_i \), we need to make a histogram using both \( \eta = \pm 1 \) in Eq. (S5).
SYMMETRY TRANSFORMATION OF ORDER PARAMETER

The O(4) order parameter discussed in the previous section $\phi_i, i = 0, 1, 2, 3$, can transform under translation operation $(T_x, T_y)$, mirror operation $(M)$, and rotation operation $(R)$ symmetry operation, we list the corresponding $4 \times 4$ group representations below.

$$T_x = \begin{pmatrix} 0 & 0 & 0 & e^{-i \frac{5\pi}{12}} \\ 0 & 0 & e^{-i \frac{5\pi}{12}} & 0 \\ 0 & e^{i \frac{5\pi}{12}} & 0 & 0 \\ e^{i \frac{5\pi}{12}} & 0 & 0 & 0 \end{pmatrix}$$  \hspace{1cm} (S17)

gives new order parameters after the configurations shift one unit along x-axis.

$$T_y = \begin{pmatrix} e^{-i \frac{\pi}{6}} & 0 & 0 & 0 \\ 0 & e^{-i \frac{5\pi}{6}} & 0 & 0 \\ 0 & 0 & e^{i \frac{\pi}{6}} & 0 \\ 0 & 0 & 0 & e^{i \frac{5\pi}{6}} \end{pmatrix}$$  \hspace{1cm} (S18)
is the transformation of order parameter after y-axis translation.

$$M = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & e^{i \frac{5\pi}{6}} & e^{i \frac{\pi}{2}} & 0 \\ e^{-i \frac{5\pi}{6}} & 0 & 0 & e^{-i \frac{7\pi}{12}} \\ e^{-i \frac{\pi}{2}} & 0 & 0 & e^{-i \frac{5\pi}{12}} \\ 0 & e^{i \frac{5\pi}{6}} & e^{i \frac{\pi}{2}} & 0 \end{pmatrix}$$  \hspace{1cm} (S19)

$M$ is a mirror operator, the mirror axis is perpendicular to the x-axis.

$$R = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & e^{i \frac{\pi}{6}} & e^{i \frac{5\pi}{6}} & 0 \\ e^{-i \frac{\pi}{6}} & 0 & 0 & e^{-i \frac{5\pi}{6}} \\ e^{-i \frac{5\pi}{6}} & 0 & 0 & e^{-i \frac{\pi}{6}} \\ 0 & e^{i \frac{\pi}{6}} & e^{i \frac{5\pi}{6}} & 0 \end{pmatrix}$$  \hspace{1cm} (S20)
is the $C_6$ rotation.

It can also be seen from above analysis that the order parameter is distributed on a four-dimensional sphere. As shown in the Fig. 3 in the main text, at the QSL-VBS transition point, the O(4) order parameter lives homogeneous on the sphere hence acquires with emergent O(4) symmetry; and deep inside the $\sqrt{12} \times \sqrt{12}$ VBS phase, the order parameters concentrate on few discrete points on the sphere, hence suggesting the O(4) symmetry breaking.