Spin nematic phases driven by the Heisenberg interactions in a spin-1 dimer system forming a bilayer

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Spin nematics had long been considered as an elusive nonmagnetic state, where the spin-1 moments lose their directions while keeping the orientations in a spatially ordered manner similar to the liquid crystals. Such order emerges as a result of transverse fluctuation of spin-1 moments, which is enhanced by the quantum many body effect. We propose a realistic model system based on dimers forming bilayers that can easily host spin nematics. Each dimer consists of antiferromagnetically coupled spin-1 pair which tend to form a dimer-singlet phase. We show that this dimer-singlet is immediately replaced with the ferroic nematic phases, when very small inter-dimer Heisenberg exchange interactions are introduced. This nematics is exotic in the sense that the spin-1 moments form a uniform Bose–Einstein condensate, whereas the nematic directors develop a spatially modulated structure. It apparently differs from the conventional ones found in a strong magnetic field or next to the ferromagnetic phases, which were often difficult to realize in experiments. Hidden nematic phases should thus exist in many of the quantum spin-dimer materials, which serve as a good platform to study nematic phases in laboratories, and a family of $\text{Ba}_3\text{ZnRu}_2\text{O}_9$ may become a first possible example.

I. INTRODUCTION

Nematics, regarded as a sort of liquid crystal in a more general context, now forms a wide range of phases of matter in crystalline solids. The “electronic nematic state” was first proposed in a doped Mott insulator as a consequence of the melting of stripes, aiming to understand the origin of high-$T_c$ phase in cuprates. More recently in iron-based superconductors, lowering of the symmetry of electronic wave functions due to the orbital ordering is regarded as a nematicity, where there exist several evidences that the fluctuation of this nematicity is related to the stability of the superconductivity particular to this system. When defined on a crystal lattice, the nematics of charges and orbitals, and also of spins are all described in a similar manner in terms of symmetry by the quadrupolar order parameter.

Spin nematics is an exotic nonmagnetic phase in insulating quantum magnets. The spin moments break their rotational symmetry, each forming a wave function in the shape of rod- or disk-like director, collectively aligned in a particular direction. The search of spin nematics has been a challenge, since experimental evidences are provided in only limited numbers of systems; in a two-dimensional layer of liquid $^3\text{He}$ in an artificially designed optical lattice, and in a quantum spin 1/2 magnets in a high magnetic field.

There had been some reasons that the quantum spin systems cannot easily become a good platform of spin nematics. In materials, the electrons carry spin-1/2, which can only form a dipole by itself, and to form a quadrupole which is a rank-2 tensor, we need at least spin-1 with three different $S^z$-levels (see Eq. (8)). There are two ways to form spin-1 from a spin-1/2. One is to use the Hund’s coupling between spin-1/2 in different orbitals on the same site, which will generate site-nematics. The other is to efficiently compose spin-1 from two spin-1/2’s on neighboring sites by the interaction, in which case the bond-nematic is formed. In the former case, one can consider as a simplest quantum spin-1 model called bilinear-biquadratic (BLBQ) model, which is known to host a spin nematic phase for a very large biquadratic interaction, $(S_i \cdot S_j)^2$, where $S_i$ is a spin-1 operator. However, the biquadratic interaction is generally much smaller than the Heisenberg interaction, so that this nematics is hardly realized in materials. To have the latter bond nematics in a spin-1/2 model, often a very high magnetic field and a frustration effect are required. In a fully polarized spin-1/2 state, the standard lowest energy excitation is an $S^z = -1$ magnon obtained after flipping a single spin-1/2 upside down. However, if there are good reasons to suppress the propagation of this magnon in space, e.g. the frustration effect on a $J_1$-$J_2$ square lattice model or a ring exchange model, the lowest excitation is replaced by the multi-magnons propagating together in space. For example, the two-magnons consist of two spin-1/2’s pointing downward, which together constitute a quadrupolar operator. They thus condense near the saturation field and form a spin nematic phase found in the spin-1/2 ladder or in liquid $^3\text{He}$. In practice, it is hard to realize such a high field in experiments, which is unfavorable for future applications.

Recently, a double-layered spin-1/2 dimer system is proposed as a platform of spin nematics in a zero magnetic field. There, the two spin-1/2’s form a singlet within a dimer by the antiferromagnetic coupling, and the inter-dimer interactions work to dope the spin-1/2 triplets. These doped spin-1’s naturally interact with each other in the same way as the BLBQ model, and two types of nematic phases are found to appear next to the singlet phase. There, the ring exchange interaction along the twisted closed path plays a key role, which cyclically permutes a set of four on-site spin-1/2’s forming two dimers. When changing the basic unit of the model to
the spin-1’s defined on dimer bonds, the original spin-1/2 ring exchange is transformed to the biquadratic interaction between spin-1’s, which supports a nematic order. Another theoretical proposal recently given is a model of ferromagnetically coupled spin-1/2 dimers, which case the spin-1 is automatically formed on a dimer bond by construction. The origin of spin nematics is similar to the above antiferro-spin-1/2 dimers; in their perturbation processes, the effect similar to the twisted ring exchange mentioned above appears. These two works show that the spin nematics is available in a much easier manner than had been believed so far. Still, in both models, it is not necessarily easy to realize the model parameters that afford spin nematics in actual material systems.

In the present paper, we show that the dimer-bond nematic phase in an antiferromagnetically coupled bilayer can be far easier realized when we replace the spin-1/2 dimer with spin-1 dimer. There are two points to be stressed; one is that the origin of the nematic phase is simply the inter-dimer Heisenberg exchange interactions. The other regarding the technical aspect is, despite a seeming difficulty in increasing the degrees of freedom of the spin moments, one can treat this model by transforming it to the effective bosonic model as a low energy approximation. This bosonic model is equivalent to the antiferromagnetically coupled spin-1/2 dimer model [9] aside from how to map the interaction parameters of spin models to those of the bosonic model. We show that a very small Heisenberg interaction between spin-1’s belonging to different dimers can generate two types of ferroic spin nematic phases, which at the same time form a Bose–Einstein condensate (BEC) of triplets. Although we deal with the dimers forming a triangular lattice, the geometrical frustration effect does not play any role in the formation of our spin nematics. Namely, one can realize similar types of spin nematics also in other geometries such as square lattice. Also, our nematics requires neither a high magnetic field nor a spin polarized ferromagnetic phase as a mother phase, and in that point, distinctively differs from the nematic magnon-bound state of the spin-1/2 systems. The present finding will thus release us from the long-standing difficulty of finding spin nematics in solids. The relevance with the actual material, Ba$_3$Mn$_2$O$_9$ ($M = \text{Zn, Ca, etc.}$), will be finally discussed.

The paper is organized as follows. In § II, we introduce the spin-1 dimer model and the transformation to the effective bosonic model. The numerical results on the bosonic model is given in § III and the origin of the spin nematics and related matters are discussed in § IV followed by a brief summary in § V.

II. DERIVATION OF THE EFFECTIVE HAMILTONIAN OF BOSONS

A. Low-energy states of spin-1 dimers

We consider a system consisting of dimers of two spin-1’s. As shown in Fig. I(a), the dimers stack parallelly and form a double-layered triangular lattice. The Hamiltonian is given as

\[ \mathcal{H} = \mathcal{H}_{\text{intra}} + \mathcal{H}_{\text{inter}}, \]

\[ \mathcal{H}_{\text{intra}} = \sum_{i=1}^{N} \left[ J S_{i_1} \cdot S_{i_2} + B (S_{i_1} \cdot S_{i_2})^2 \right], \]

\[ \mathcal{H}_{\text{inter}} = \sum_{\langle ij \rangle} \sum_{\gamma=1,2} (J' S_{i_\gamma} \cdot S_{j_\gamma} + J'' S_{i_\gamma} \cdot S_{j_\gamma}). \]

where $S_{i_\gamma}$ is the spin-1 operator of $\gamma$-th site on an $i$-th dimer. The summation $\langle ij \rangle$ is taken over all the neighboring pairs of dimers, and $1 = 2$ and $2 = 1$. $J$ ($> 0$) and $B$ ($> 0$) denote the antiferromagnetic Heisenberg and the biquadratic interactions, respectively, $J'$ and $J''$ are the inter-dimer Heisenberg interactions (Fig. II(b)), and $N$ is the number of dimers. We analyze the Hamiltonian Eq. (1) by first transforming it to the effective model of the spin-1 bosons via the perturbation theory, and then solving it by the numerical diagonalization on a finite cluster.

Let us first consider an isolated dimer consisting of two spin-1 interacting via the BLBQ interactions $\mathcal{H}_{\text{BLBQ}} = J S_1 \cdot S_2 + B (S_1 \cdot S_2)^2$. The energy eigenstates of $\mathcal{H}_{\text{BLBQ}}$ are classified into singlet ($s$), triplets ($t$), and quintets ($q$), and their energies are given as $e(s) = -2J + 4B$, $e(t) = -J + B$, and $e(q) = J + B$, respectively. Figure III(c) shows these energy levels as a function of $B/J$. At small $B/J$, the Heisenberg interaction is dominant and the lowest energy state is a singlet. This singlet state is replaced by the triplet state when $B/J > 1/3$, while the quintet cannot have lower energy than the triplet and remain as the excited states.

As a starting point of the perturbation, we take $\mathcal{H}_{\text{inter}} = 0$, where the ground state is the product state of the singlets on the isolated dimers for $B/J < 1/3$, and that of triplets for $B/J > 1/3$. In introducing $\mathcal{H}_{\text{inter}} \neq 0$, we consider the processes up to second order in $J'/J$ and $J''/J$, so that the effective interactions between two adjacent dimers appear mainly in the result. The energies of the disconnected two dimers with $\alpha$- and $\beta$-multiplets $E(\alpha, \beta)$ are shown in Fig. III(d). One can see that the states including quintets are higher in energy than the states without quintets when $B/J < 2/3$. Therefore, based on the natural assumption that $B/J$ is small enough, we construct the effective Hamiltonian for the low-energy manifold of states including only singlets and triplets.

The first order process contributes to the energy correction of singlet and triplet states, as well as to the exchange of triplet and singlet on the neighboring two
dimers. Within the second order perturbation processes between two adjacent dimers, the intermediate excited states have at least one quintet as shown in the examples of the processes; in Fig. 1(e), the two-dimer state \(|s, t_0\rangle\) returns to the same state through the excited states \(|t_0, q_0\rangle, |t_1, q_{-1}\rangle\) and \(|t_{-1}, q_{+1}\rangle\), where \(s\) is the singlet state, and \(|t_0\rangle\) and \(|q_0\rangle\) are the triplet and the quintet states with \(S^z = \mu\), respectively. In the processes shown in Fig. 1(f), \(|t_{+1}, t_{-1}\rangle\), the two-dimer states with \(S^z = +1\) and \(S^z = -1\) triplet dimers, mixes with \(|t_{-1}, t_{+1}\rangle\) via the three excited states \(|s_0, q_0\rangle, |q_0, s_0\rangle\) and \(|q_0, q_0\rangle\).

The low-energy basis can be described in the spin-1 hard-core bosonic language. The singlet corresponds to the vacuum, and the triplets are the bosons which are not allowed to doubly occupy a dimer. This kind of treatment is equivalent to the bond-operator approach, developed for the spin-1/2 dimer system [40,41], and later applied to spin-1 dimer system [42,43] and also to general spin-S dimer [44]. We choose the time-reversal invariant form of the basis set \(||t_{i,\alpha}\rangle\rangle\) described as,

\[
|t_{i,x}\rangle = \frac{i}{\sqrt{2}} (|+1, 0\rangle - |0, 1\rangle),
\]

\[
|t_{i,y}\rangle = \frac{i}{\sqrt{2}} (|+1, 0\rangle - |0, -1\rangle),
\]

\[
|t_{i,z}\rangle = -i (|+1, -1\rangle - |0, +1\rangle),
\]

where the dimer states on the r.h.s. described as \(|S^z_{t_1}, S^z_{t_2}\rangle\) are those classified by the \(S^z\)-values of the two spins forming a dimer. The details of the bond-operator approach and the description of the original spin operators using the bosonic operators are shown in Appendix A1.

### B. Effective Hamiltonian

The triplet state with \(\alpha\)-component \(|t_{i,\alpha}\rangle\) at site-\(i\) is expressed as \(b^\dagger_{i,\alpha}|0\rangle\), where \(|0\rangle\) is the singlet state and \(b^\dagger_{i,\alpha}\) is the creation operator of a boson representing that triplet. Using this bosonic operator, the effective Hamiltonian \(H_{\text{eff}}\) up to second order in \(J'/J\) and \(J''/J\) is given as

\[
H_{\text{eff}} = E_0 + H_\mu + H_t + H_P + H_V + H_J + H_B + H_{3\text{body}},
\]

where \(E_0\) is the energy of the ground state of the original Hamiltonian \(H\), \(H_\mu\) is the hard-core condition \(n_i = 0\) on site-\(i\), \(H_t\) is the parameter included in \(H_{\text{eff}}\) as expressed by the singlet \(|S^z = +1\rangle\), \(H_P\) is the parameter included in \(H_{\text{eff}}\) as expressed by the triplet \(|S^z = 0\rangle\), \(H_V\) is the parameter included in \(H_{\text{eff}}\) as expressed by the quintet \(|S^z = -1\rangle\), \(H_J\) is the parameter included in \(H_{\text{eff}}\) as expressed by the singlet \(|S^z = +1\rangle\) and \(|S^z = 0\rangle\) and \(|S^z = -1\rangle\), \(H_B\) is the parameter included in \(H_{\text{eff}}\) as expressed by the singlet \(|S^z = +1\rangle\) and \(|S^z = 0\rangle\) and \(|S^z = -1\rangle\), and \(H_{3\text{body}}\) is the parameter included in \(H_{\text{eff}}\) as expressed by the singlet \(|S^z = +1\rangle\) and \(|S^z = 0\rangle\) and \(|S^z = -1\rangle\).

The parameters included in \(H_{\text{eff}}\) are described by the original interaction parameters in Eq. (1) as

\[
E_0 = (2 J + 4 B) N,
\]

\[
\mu = -J + 3B + \frac{20 z}{27 (J - B)} (J' - J'')^2,
\]

\[
t = \frac{4}{3} (J' - J''),
\]

\[
P = -\frac{4}{3} (J' - J''),
\]

\[
V = \left[ -\frac{40}{27 (J - B)} - \frac{8}{9 (J + 3B)} - \frac{2}{9 J} \right] (J' - J'')^2,
\]

\[
J = \frac{J' + J''}{2} + \left[ -\frac{16}{9 (J + 3B)} + \frac{1}{12 J} \right] (J' - J'')^2,
\]

\[
B = \left[ -\frac{4}{9 (J + 3B)} - \frac{1}{144 J} \right] (J' - J'')^2,
\]

where \(z\) is the coordination number. One can immediately see that \(\mu, t, P\) and \(J\)-terms include the terms that originate from the first order process, whereas \(V\) and \(B\)-terms do not.

There are some processes at the second order level where the three dimers take part in, which we denote as \(H_{3\text{body}}\) in Eq. (3). We numerically evaluate the effects of \(H_{3\text{body}}\) on the effective Hamiltonian by comparing the energies of the ground states of the original Hamiltonian \(H\) (Eq. (1)), and of the effective Hamiltonian \(H_{\text{eff}}\) (Eq. (3)) with and without \(H_{3\text{body}}\) in a small cluster, finding that it does not play a significant role. We thus discard this \(H_{3\text{body}}\) term in the following for simplicity. The details of the evaluation of the effective model is shown in the Appendix A2. We further show that even the other second order terms included in Eq. (3), do not contribute...
much to the majority of phases we deal with. The way how the inter-dimer interactions work thus turns out to be surprisingly simple.

C. Physical quantities

For the analysis of the effective model, we calculate the following properties that characterize the ground state. The boson density per dimer is denoted as \( \langle n_i \rangle = N^{-1} \sum_{i=1}^{N} \langle n_i \rangle \), and its structure factor is given as

\[
N(k) = \frac{1}{N} \sum_{i,j=1}^{N} \langle n_i n_j \rangle e^{i \mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)}.
\]  

(5)

The magnetic properties are examined by the spin and quadrupole structure factors

\[
S(k) = \frac{1}{N} \sum_{i,j=1}^{N} \langle \mathbf{S}_i \cdot \mathbf{S}_j n_i n_j \rangle e^{i \mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)},
\]

(6)

\[
Q(k) = \frac{1}{N} \sum_{i,j=1}^{N} \langle \mathbf{Q}_i \cdot \mathbf{Q}_j n_i n_j \rangle e^{i \mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)},
\]

(7)

where \( \mathbf{Q} \) is the 5-component vector representation of quadrupole operator of spin-1 bosons defined as

\[
\mathbf{Q}_i = \begin{pmatrix}
Q_i^{x^2-y^2} \\
Q_i^{x^2-z^2-r^2} \\
Q_i^{xy} \\
Q_i^{yz} \\
Q_i^{zx}
\end{pmatrix} = \begin{pmatrix}
( S_i^x )^2 - ( S_i^y )^2 \\
\frac{1}{\sqrt{3}} [ 3 ( S_i^z )^2 - S( S + 1) ] \\
S_i^x S_i^y + S_i^y S_i^z + S_i^z S_i^x \\
S_i^y S_i^z + S_i^z S_i^x + S_i^x S_i^y \\
S_i^x S_i^y + S_i^y S_i^z + S_i^z S_i^x
\end{pmatrix}.
\]  

(8)

In a system with spin-1 defined on each site, typically represented by the spin-1 BLBQ models, the quadrupole operator \( \mathbf{Q} \) is the on-“site” operator. For a system with spin-1/2 per site, the quadrupole operator is defined on a bond instead, since one needs to prepare a spin-1 from spin-1/2 per site, the quadrupole operator is defined on the quadrupolar states.

III. RESULTS

A. Phase diagram

We numerically diagonalize \( \mathcal{H}_{\text{eff}} \) on the \( N = 12 \) triangular lattice (\( z = 6 \)) under the periodic boundary condition. The phase diagrams on the plane of \( J' / J \) and \( J'' / J \) at \( B / J = 0.2 \) and 0.4 are shown in Figs. 2(a) and 2(b).

The phase diagram is divided into four parts in overall. When \( J' \sim J'' > 0 \), the antiferromagnetic phases with \( \langle n_i \rangle \approx 1 \) (AFM-solid) and \( \langle n_i \rangle \lesssim 0.9 \) (AFM-BEC) are stabilized by the antiferromagnetic interaction, \( J' > 0 \), between bosons occupying the neighboring dimers. On the opposite part of the phase diagram, \( J' \sim J'' < 0 \), the ferromagnetic phase with \( \langle n_i \rangle \approx 1 \) (FM-solid) is realized for the similar reasons. When \( J' \sim J'' < 0 \) and \( J' - J'' > 0 \), two different types of spin nematic (ferroquadrupole (FQ)) phases, FQ-BEC-1 and FQ-BEC-2 appear over a wide parameter region. Throughout both of the phase diagrams, we see no particular features of bosons, i.e., \( N(k) \) takes the maximum value at \( \Gamma \)-point, which indicates that bosons distribute uniformly in space and does not show any translational symmetry breaking long range order.

B. \( J' = J'' \) line

The starting point is \( J' = J'' = 0 \), at which the ground state is the product state of the isolated-dimer state. As one can see from Eq. [1], most of the parameters, namely \( t, P, V, B \) are the linear or the square functions of \( (J' - J'') \). Therefore, these parameters remain zero exactly at \( J' = J'' \), namely the inter-dimer interactions cancel out because of the geometrical frustration effect.
In fact, when $B/J = 0.2$, the singlet product state, namely $(n_i) = 0$, remains a ground state along this line. The endpoint of this singlet phase is evaluated in the following manner; when $J' = J''$, the effective Hamiltonian consists only of two terms

$$\mathcal{H}_{J' = J''} = -\mu \sum_{i=1}^{N} n_i + J \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j n_i n_j,$$  

(13)

with $\mu = -J + 3B$ and $J = J'$. Regardless of its sign, $J$ works as an effective attractive interaction between bosons, since it is energetically favorable to occupy the neighboring pairs of dimers with triplets to gain the magnetic interaction energy. Then, there is a first order transition between the $\langle n_i \rangle = 0$-singlet and the $\langle n_i \rangle = 1$-FM or AFM solid phases. The phase boundary can be obtained by comparing their energies, $E_0(N)$ and $E_1(N)$, where there is a relationship,

$$E_1(N) = E_0(N) - \mu N + 3N e_{\text{bond}}.$$  

(14)

Here, $e_{\text{bond}}$ is evaluated as the bond-energy of the ground state of the spin-1 triangular lattice Heisenberg model $J' \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j$ for $N = 12$. Figure 2(c) shows the resultant phase diagram on the plane of $J' = J''$ and $B$ with $J = 1$. The singlet phase corresponding to the straight line in Fig. 2(a) shrinks toward smaller $J' = J''$ value with increasing $B/J$, and disappears at $B/J = 1/3$. For $B/J > 1/3$, $\langle n_i \rangle = 1$ is realized throughout the whole $J' = J''$ line.

C. Ferromagnetic and antiferromagnetic phases

The FM and AFM phases extend from the endpoints of the $J' = J''$-singlet phase discussed above. In Fig. 3 we show the total energy $e_{\text{total}}$ and the contributions from each terms, $e_t$, $e_r$, $e_J$, and $e_B$, the boson density, $\langle n_i \rangle$, and the values of the structure factors at $\Gamma$, $K$ and M points of the Brillouin zone. We vary $J'/J$ along the fixed $J''/J = -0.2$ and 0.1 lines. In the former case, a jump in the physical quantity is found at the transition from the FM-solid to the FQ-BEC-2 phase. Compared to other phases, the FM-solid phase has a large energy gain of $e_J$, indicating that the magnetic interaction $J$ is responsible for stabilizing the FM-solid. Indeed, $S(k)$ shows a peak at the $\Gamma$-point in this phase while the other $Q(k)$ and $\langle Q(k) \rangle$ remain small. When we vary $J'/J$ along $J''/J = 0.1$ (Figs. 3(d)-(f)), $\langle n_i \rangle \lesssim 0.55$, and the system remains a BEC. The transitions along this line are of second order. At $J'/J \gtrsim 0.3$, $S(k)$ at K-point starts to overwhelm $Q(k)$ at the $\Gamma$-point which we recognize as the AFM-BEC phase, following the treatment in Ref. 19 (see the last part of §1C). The phase boundaries in Fig. 2 are classified into first and second order ones (filled and open circles) according to this analysis.

D. Spin nematic phases

In the phase diagram, there are two different spin nematic phases, FQ-BEC-1 extending at $J'' > J'$ and FQ-BEC-2 at $J'' < J'$. As we see in Figs. 3(e) and 3(f), $\langle n_i \rangle$ and $Q(k)$ both once decrease down to zero at the boundary of the two phases where the singlet state appears, which marks the second order transition. In such a case, the order parameters of the two phases should differ. In fact, although $Q(k)$ at $\Gamma$-point is dominant in both phases, only in the FQ-BEC-2 phase $Q(k)$ at K-point takes as large value as well.

Figures 4(a) and 4(b) show the two-point quadrupole correlations between site-1 and site-$j$, $(\mathbf{Q}_1 \cdot \mathbf{Q}_j)$, in FQ-BEC-1 and FQ-BEC-2 phases. The former correlation develops uniformly in space, whereas in the latter, there is apparently a growth of correlation in the period of twice the lattice spacing in all three directions. This three-sublattice-like structure of quadrupole moments corresponds to the peak of $Q(k)$ at the K-point. Figures 4(c) and 4(d) are the two-point correlation of bosons, $\langle n_i n_j \rangle$, which are both uniform in space. This indicates that the three-sublattice structure of the quadrupolar moment in the FQ-BEC-2 is not because of the modulated the bosonic distribution but originates purely from the correlation between the spin degrees of freedom $\mathbf{S}_j$: the nearest-neighbor quadrupolar correlation is suppressed, while the next nearest neighboring correlations are ferroic.

E. Case of $B/J = 0.4$

We now focus on the case of $B/J = 0.4$, where $\mu$ takes a positive value. The singlet phase no longer exists and the triplet product state realized at $J' = J'' = 0$ immediately transforms to either of the phases we discussed earlier when the inter-dimer interactions become finite. Figure 5 shows the $J'/J$ dependences of energies, boson density, and the structure factors to be compared with Fig. 3. The first order transitions separating the FM-solid from FQ phases are observed. The boson density in the FQ-BEC phase remains quite stable at around $\langle n_i \rangle \approx 0.55$, indicating that the nature of the BEC phases does not change much with $B/J$.

F. The orders of perturbation

The interaction parameters in Eq. (4) include the first and second order terms. Among them, $V$ and $\mathcal{B}$ disappear when we neglect the second order terms. To see how much the second order terms contribute to the determination of the phase diagram, we perform the numerical diagonalization by limiting the parameter values to those up to the first order in $J'/J$ and $J''/J$ with $N = 12$. Figures 6(a) and 6(b) show the phase diagrams at $B/J = 0.2$ and 0.4. The phase diagrams are in good
agreement with those in Figs. 2(a) and (b), indicating that \( V \) and \( B \) do not play a major role in the five representative phases, FM-solid, AFM-solid/BEC, FQ-BEC-1 and FQ-BEC-2.

IV. DISCUSSION

In spin-1 BLBQ models, the spin nematic phase appears when \(|B| \gtrsim |J|\). This is because the model is written in the form,

\[
JS_i \cdot S_j + B(S_i \cdot S_j)^2 = \frac{2J - B}{2}S_i \cdot S_j + \frac{B}{2}Q_i \cdot Q_j + \text{const.} \tag{15}
\]

which explicitly shows that the dipolar (magnetic) and quadrupolar orders compete with each other, and the latter appears when the latter term overwhelms the former. However, in our effective model (Eq. (3)), the biquadratic interactions \( B \) only appears at the second order level, and indeed, the results in Fig. 6 show that it does not play a role to stabilize our FQ phases. One can also confirm that is the K-point for \( J' = J'' < 0 \) and \( J' - J'' > 0 \), respectively, at \( B/J = 0.2 \). When the bottom of the band touches the zero level, the instability takes place and the \( \beta_{k,\alpha} \)-bosons of that wave number condense and form a BEC phase. This happens by increasing \( J' = -J'' \) only up to \( |J' - J''| \sim 0.05 \), which is consistent with the numerical analysis that the singlet product state immediately gives way to the FQ phases in the \( J' = -J'' \) direction. The wavenumber at which the \( \epsilon_k \) takes the minimum is the \( \Gamma \)-point when \( J' - J'' < 0 \), whereas it is the K-point for \( J' - J'' > 0 \). The former is the usual uniform FQ ordering, and the latter explains well the particular three-sublattice-like structure of the quadrupole correlations in FQ-BEC-2 phase we saw in Fig. 7(b).

We would like to note that the above analysis is different from the instability analysis performed for a nematic order based on a magnon-pair condensation in the spin-1/2 \( J_1-J_2 \) model. In these cases, one-magnon and two-magnon instabilities from the fully polarized ferromagnetic phase in a high magnetic field are examined. Because of the frustration effect, the kinetic motion of a single magnon is prohibited, while instead the two magnons form a bound state and propagate in space via two-body pair hopping term, which is the origin of the nematic order by definition, since a pair of spin-1 bosons, \( \langle b_i^\dagger b_j \rangle = \langle S_i^- S_j^- \rangle = Q e^{2\theta} \neq 0 \), is the quadrupolar order parameter itself. In our case, a one-body pair creation/annihilation term is the source of the instability, and a one-boson branch condenses and form the FQ phases, which is an off-diagonal long range order of three-fold SU(2) bosons \( \langle \beta_{k,\alpha} \rangle \neq 0, \alpha = x, y, z \).

Previously, in a spin-1/2 dimer system, we showed that the origin of the nematic phase is the ring exchange interactions that permutate the four spin-1/2 along the twisted path as, \((1, 2, 3, 4) \rightarrow (2, 3, 4, 1)\), which is shown in Fig. 8(a). In that case, the two spin-1/2’s on a dimer form an \( S_i = 1 \) triplet, and the ring exchange interaction exchanges the spin-1’s on neighboring dimers, \( \langle S_i^z S_j^z \rangle = \langle +1, -1 \rangle \) with \(( -1, +1 \rangle \)-states (see Fig. 8(b)). This plays the same role as the biquadratic interaction, \( B(S_i \cdot S_j)^2 \), and when all the dimers are filled with a triplet, \( S_i = 1 \), the system is reduced to the BLBQ model. When the dimers are not fully occupied with triplets, there appears another nematic phase consisting of spin-1’s forming BEC, which is very similar to the FQ-BEC-2 phase in the present work. However, since the magnitude
of the ring exchange interactions required is about 1/5 of that of the intra-dimer Heisenberg interaction, it is not always possible to realize these nematic phases.

Another spin-1/2 dimer model with ferromagnetic intra-dimer coupling is recently proposed. They showed that the exchange interaction, $J'$ and $J''$ ($J_1$ and $J_2$ in their notation), operated twice at the second order perturbation is important to stabilize the nematic phase. As shown schematically in Fig. (8a), this works in the same manner as the ring exchange interaction, and generates an effective biquadratic term. However, this time they need a larger $J''$ as their nematics need to compete with the stable ferromagnetic phase.

In our spin-1 dimer, the pair-creation and annihilation term results in an off-diagonal pair condensation of up and down spin-1’s via the processes shown in Fig. (8c). These processes, when performed twice, will give the same effect as the biquadratic interaction in Fig. (8b). The advantage here is that it is a first order process and can be more easily realized than $\mathcal{B}$ or the ring exchange processes.

We finally discuss the relevance with the actual material. In a family of Ba$_3$M$_2$Ru$_2$O$_8$ ($M = \text{Ca}, \text{Zn}, \text{Ni}, \text{Co, Sr}$) the two face-shared RuO$_6$ octahedra form a dimer which is stacked along the two-dimensional triangular lattice in the same way as our model Fig. (1a). The Ru$^{5+}$ carries either $S = 3/2$ or $S = 1$ and the material is well described by our Hamiltonian, Eq. (1). Notice that even for $S = 3/2$, the system is reduced to the same effective model, Eq. (3). Intriguing magnetic properties were reported; For a Zn-compound, the uniform susceptibility is strongly suppressed down to 32 mK, a much lower temperature than the value of $J \sim 150 - 200$ K. In a Co- or Ni-compound with shorter inter-dimer distances, namely having larger $|J'/J|$ and $|J''/J|$, the system undergoes an antiferromagnetic transition at $T_N \sim 100$ K. The Ca- and Sr-compound with the longer inter-dimer distances contrarily favor a standard nonmagnetic singlet state. Usually, $J, J'$ and $J''$ are antiferromagnetic ones, and if we increase the inter-dimer interactions from the center of the phase diagram in Eq. (3) toward the upper right direction, the ground state transforms from singlet, FQ-BEC-2, and to an antiferromagnetic phase, in good agreement with the experimental observation, (Sr, Ca)$\rightarrow$(Zn)$\rightarrow$(Co, Ni). Since the spin nematic phases conventionally discussed were all found next to the fully polarized ferromagnetic/antiferromagnetic phase, the exotic nonmagnetic phase in the Zn-compound was not really connected to the spin nematics. Our series of studies on spin dimer systems are the first to point out that the spin nematics which is the physics of spin-1 can live next to the spin-0 singlet phase. It is difficult to experimentally identify the thermodynamic spin nematic phase, since this kind of symmetry breaking is not detected from the standard susceptibility measurements. There should thus be a hidden spin nematics in numbers of spin dimer systems still unexplored.

V. SUMMARY AND PERSPECTIVE

In conclusion, we found two different types of ferroic spin nematic BEC phases in the spin-1 dimer model. In this model, the dimerized two spin-1’s form a bilayer triangular lattice, and the antiferromagnetically coupled dimer-spins tend to form a bond-singlet. Starting from the limit of singlet phase without inter-dimer interactions, and including the inter-dimer Heisenberg exchange terms $J'$ and $J''$, perturbatively up to second order, we derived an effective hard-core bosonic model describing the low-energy properties of the original model. The lowest energy state of the unperturbed Hamiltonian at $B/J < 1/3$ is the product state of the dimer singlets, which is considered as a vacuum in the bosonic model, and the inter-dimer interactions work to dope the spin-1 bosons in the excited states. The major part of the bosonic Hamiltonian consists of the hopping ($t$) and pair-creation and annihilation ($P$) of bosons, as well as the chemical potential ($\mu$) and the antiferromagnetic exchange interaction ($J$). The bosons are doped by $\mu$ and the $t$-term contributes to the formation of BEC. The ferromagnetic (FM) and antiferromagnetic (AFM) phases appear due to $J < 0$ and $J > 0$. We find two counterparts of these magnetic phases, which are the FQ-BEC-1 and FQ-BEC-2 phases that are found to appear due to $P$. In both of the FQ phases, the boson density takes the value $\approx 0.55$, forming BEC, and show ferro-quadrupolar (FQ) correlations. The former has the uniform FQ-correlations, whereas the latter has the three-sublattice-like FQ structures although the bosons distribute uniformly in space. Since $P$ works as a density fluctuation of bosons, the bosons condense into a FQ-BEC, which is the off-diagonal long range order of both spin-1 and bosonic operators.

Our results are widely applied to the bilayer quantum spin dimer systems, since the interactions appear in Eq. (4), are all standard ones that are derived naturally from the strong coupling perturbation theory of Mott insulator; the Heisenberg exchange interactions, $J, J'$ and $J''$, and the biquadratic intra-dimer interaction $B$. Although we did not discuss much the details, the physics presented is not much influenced by the magnitude of $B$, and the value of $B$ is reported to be relatively larger than it has been believed before. The FQ-BEC phases discussed here are exotic in the sense that the distribution of $S = 1$-bosons are uniform in space, whereas the quadrupolar correlation may develop a particular spatial modulation. It differs from the nematic solid phases of the $S = 1$ BLQ model, and from the magnon-bound states of the $S = 1/2$ models in high fields. It may thus form a new class of spin nematic BEC phase, ubiquitous in quantum spin dimer systems to be explored in laboratories.
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Appendix A: Details on the effective model Eq. (3)

We show the details of the derivation and the reliability of the effective hard-core boson model shown in Eqs. (3) and (4).

1. Construction of a time-reversal invariant basis of spin-1 dimer state via bond-operator approach

To make direct connections of our representation in the main text with the previous studies, we introduce the frequently used bond-operator representation of the spin-1 state in a unit of dimer, where we adopt the one that keeps the time-reversal symmetry. Notice that the basis states used in the previous studies on the bond-operator approach\[22,23] break the time-reversal symmetry.

First, we show the details of the derivation and the reliability of the effective hard-core boson model shown in Eqs. (3) and (4).

For the singlet state, we set

$$\left| s \right>=\frac{1}{\sqrt{3}}\left| x,x \right>+\left| y,y \right>+\left| z,z \right>$$

(A2)

for the singlet state,

$$|t_{i,\alpha}\rangle=\frac{1}{\sqrt{2}}\sum_{\beta,\gamma}\varepsilon_{\alpha\beta\gamma}|\beta,\gamma\rangle$$

(A3)

for the triplet states, and

$$|q_{i,\alpha\beta}\rangle=-\frac{1}{\sqrt{2}}\left(|\alpha,\beta\rangle+|\beta,\alpha\rangle\right)+\left(\sqrt{2}-1\right)\delta_{\alpha\beta}|\alpha,\alpha\rangle$$

(A4)

for the quintet states. Since only three of four states $|s\rangle$, $|t_{i,\alpha}\rangle$, $|q_{3a^2-r^2}\rangle$, and $|q_{3b^2-r^2}\rangle$ are linearly independent, we adopt $|s\rangle$ and a pair of states $|q_{3a^2-r^2}\rangle$ and $|q_{3b^2-r^2}\rangle$ with $\beta,\gamma \neq \alpha$ as the basis states. For $|g_{3a^2-r^2}\rangle$ and $|g_{3b^2-r^2}\rangle$, we choose the following representation that has good correspondence with the $S_{i,\alpha}^\mu$ operators,

$$|g_{3a^2-r^2}\rangle=-\frac{1}{\sqrt{6}}\left(2|x,z\rangle-|x,x\rangle-|y,y\rangle\right),$$

$$|g_{3b^2-r^2}\rangle=-\frac{1}{\sqrt{2}}\left(|x,x\rangle-|y,y\rangle\right).$$

(A5)

In the main text, we adopted the singlet state as a vacuum, whereas in this bond-operator approach, we redefine the vacuum as the state without any multiplet. Accordingly, instead of $b_{i,\alpha}$ and $b_{i,\alpha}^\dagger$, we use $s_{i}$ $(s_{i}^\dagger)$ and $t_{i,\alpha}$ $(t_{i,\alpha}^\dagger)$ as the annihilation (creation) operators of the singlet and triplet of component-$\alpha$. Then, the spin-1 operator $S_{i,\alpha}^\mu$ $(\mu = 1, 2, 3)$ in the $i$-th dimer can be written as follows;

$$S_{i,\alpha}^1=\frac{1}{\sqrt{3}}\left(t_{i,\alpha}^\dagger s_{i}-s_{i}t_{i,\alpha}\right)\frac{1}{2}\sum_{\beta,\gamma}\varepsilon_{\alpha\beta\gamma}t_{i,\beta}^\dagger t_{i,\gamma}$$

$$-\frac{1}{\sqrt{3}}\left(q_{i,3a^2-r^2}^\dagger t_{i,\alpha}-t_{i,\alpha}^\dagger q_{i,3a^2-r^2}\right)$$

$$-\frac{1}{2}\sum_{\beta\neq\alpha}\left(q_{i,\alpha\beta}^\dagger t_{i,\beta}-t_{i,\beta}^\dagger q_{i,\alpha\beta}\right)-\frac{1}{2}\sum_{\beta,\gamma}\varepsilon_{\alpha\beta\gamma}q_{i,\alpha\beta}^\dagger q_{i,\beta\gamma}$$

$$-\frac{1}{2}\sum_{\beta,\gamma}\varepsilon_{\alpha\beta\gamma}\left(q_{i,\alpha\beta}^\dagger q_{i,\beta\gamma}^\dagger q_{i,\beta\gamma}q_{i,\beta\gamma}-q_{i,\beta\gamma}^\dagger q_{i,\beta\gamma}q_{i,\beta\gamma}^\dagger\right).$$

(A6)

2. Evaluation of the effective model

We examine the effect of the three-dimer interactions $H_{3\text{body}}$ in the effective Hamiltonian $H_{\text{eff}}$ (Eq. (3)), which was discarded in the calculation of Figs. 2-4 in the main text.

First, we show some details of $H_{3\text{body}}$, which originates from the three-dimer processes at the second order of perturbation. We show two examples of these processes in Figs. 9(a) and 9(b), where $s$, $t$, $q$ are the singlet, triplet, and quintet states, respectively, on a dimer. Figure 9(a) is the processes similar to the correlated hoppings found in the Shastry–Sutherland model\[22,23] and Fig. 9(b) is the pair-creation of bosons.

In treating these three-dimer processes, we examined the validity of restricting the low-energy manifold of states to those including only singlet and triplets. Figure 9(c) shows the energy diagram of the three-dimer states, $E(\alpha,\beta,\gamma)$ $(\alpha,\beta,\gamma = s, t, q)$. We see that $(t, t, t)$ states and $(s, s, q)$ states are degenerate at $B/J = 0$, whereas they are well separated when a small $B/J > 0$ is introduced.

Next, we compare the ground state energies of the effective model $H_{\text{eff}}$ (Eq. (3)) with and without $H_{3\text{body}}$, and the energy of the original spin-1 dimer model $H$ (Eq. (1)). We used the 9-dimer triangular lattice under...
the periodic boundary condition. The cases of $B/J = 0.2$ are shown in Figs. 9(d) and 9(e), and those of $B/J = 0.4$ are in Figs. 9(f) and 9(g), where the parameters are chosen as $(J' + J'')/J = +0.2$ and $-0.2$. It is confirmed that the energies of $H_{\text{eff}}$ with $H_{\text{3dimer}}$ are not always closer to those of $H$ than those of $H_{\text{eff}}$ without $H_{\text{3dimer}}$, although $H_{\text{eff}}$ fully takes the second order perturbation terms into account. We see that for $|J'/J|$ and $|J''/J| \lesssim 0.2$, the energies are in good consistency with each other. The effective model may not hold quantitatively when either of $J'$, $J''$ has a large value. This might be because the three-dimer interactions appear the higher order of $J'$ and $J''$, which would cancel out the three-dimer interactions derived at the 2nd order. As we already saw in §[17], the effect of second order perturbation is small, and setting $H_{\text{body}} = 0$ does not change both the quantitative and qualitative aspects of the results. The advantage of having a simple Hamiltonian, $H_{\text{eff}}$, is that it corresponds exactly to the spin-1/2 model, and resultantly, the two models of different spin numbers can be compared on equal ground.

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spin moment in RuO$_6$ still has an ambiguity between $S = 3/2$ or $S = 1$, since the energy splittings depend on the ligands, whose details are not clarified.

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FIG. 1. (a) Schematic illustration of the spin-1 dimer model on a triangular lattice. (b) Intra- and inter-dimer interactions, where $J > 0$, $J'$, $J''$ are the Heisenberg exchanges, and $B$ is the biquadratic exchange. (c) Eigenenergy levels of $\mathcal{H}_{\text{intra}}$ of an isolated dimer. $s$, $t$, and $q$ denote the singlet, triplet, and quintet states of the dimer, respectively. (d) Eigenenergy levels of $\mathcal{H}_{\text{intra}}$ of two isolated dimers without the inter-dimer $J'$ and $J''$. (e), (f) Examples of the second order perturbation processes. Processes in panel (e) returns to the original state $|s, t_0\rangle$ and those in panel (f) exchange the $S^z = +1$ and $S^z = -1$ triplets. They are the origins of the biquadratic interactions between two triplets $(S_i \cdot S_j)^2$. 
FIG. 2. Ground state phase diagram of the spin-1 dimer triangular lattice at (a) $B/J = 0.2$ and (b) 0.4, obtained by the numerical diagonalization of the effective model with $N = 12$. Filled and open circles represent the first and second order phase transitions, where the transition between FM-BEC and FQ-BEC-2 is weakly first order. FM, AFM, FQ represent the ferromagnetic, antiferromagnetic and ferro-quadrupolar phases, and $\langle n_t \rangle \approx 1$ and $\lesssim 0.9$ are the solid and BEC states of bosons. Colors in the phase diagram are the density plot of the triplet number $\langle n_t \rangle$. (c) Phase diagram on the plane of $J'/J$ and $B/J$, whose fixed $B/J$-lines correspond to the $J' = J''$ line of the phase diagrams in panels (a) and (b).
$B/J = 0.2$

(a) $J''/J = -0.2$

(b) $J''/J = +0.1$

FIG. 3. $J'/J$ dependences of the physical quantities at $B/J = 0.2$ when (a)-(c) $J''/J = -0.2$ and (d)-(f) $J''/J = +0.1$. (a), (d) Total energies $e_{\text{all}}$ and the contributions from major terms in the effective Hamiltonian, $e_t$, $e_P$, $e_J$ and $e_B$. (b), (e) Triplet densities $\langle n_t \rangle$. (c), (f) Spin (Eq. (6)) and quadrupole (Eq. (7)) structure factors at $\Gamma$, $K$, $M$-points in reciprocal space. Quadrupole structure factors denoted as $\bar{Q}(k)$ are normalized to be compared with the spin structure factors $S(k)$. 
FIG. 4. Spatial correlation functions for \( J'/J, J''/J \) = \((-0.2, +0.2)\) in FQ-1 and \((+0.2, -0.2)\) in FQ-2 phases at \( B/J = 0.2 \); the quadrupolar correlations in (a), (b) \( \langle Q_1 \cdot Q_j \rangle \) and the boson-boson correlation in (c), (d) \( \langle n_1 n_j \rangle \). Areas of the circles are proportional to the amplitude of correlations, \( |\langle Q_1 \cdot Q_j \rangle| \) or \( |\langle n_1 n_j \rangle| \). Red and blue circles in (a), (b) correspond to the signs of \( \langle Q_1 \cdot Q_j \rangle \), positive and negative, respectively.
FIG. 5. $J'/J$ dependence of the physical quantities at $B/J = 0.4$ when (a)–(c) $J''/J = -0.1$ and (d)–(f) $J''/J = +0.2$. (a), (d) Total energies and partial energies of some terms of the effective Hamiltonian. (b), (e) Triplet densities. (c), (f) Spin (Eq. (6)) and quadrupole (Eq. (7)) structure factors of some points in the reciprocal space. The quadrupole structure factors are normalized to $\bar{Q}(k)$ to be compared with the spin structure factors $S(k)$.

FIG. 6. (a), (b) Phase diagrams of the effective model up to the first order in $J'/J$ and $J''/J$ at (a) $B/J = 0.2$ and (b) $B/J = 0.4$, obtained by the analysis of the results of the numerical diagonalization on the $N = 12$ cluster.
FIG. 7. Energy bands of the eigenstates of the quadratic Hamiltonian (Eq. (16)) for (a) \( J' - J'' < 0 \) and (b) \( J' - J'' > 0 \). We set \( B/J = 0.2 \), and use the parameters \( t, P \) and \( \mu \) defined in Eq. (4), at the first order.

FIG. 8. Three different types of fluctuations that contribute to the formation of the spin nematics. Single and double arrows represent the spin-1/2 and spin-1, respectively. (a) In spin-1/2 dimer system, ring exchange interaction that permutes spin-1/2’s as \((1, 2, 3, 4) \leftrightarrow (2, 3, 4, 1)\) in the upper panel (see Refs. 30 and 38) and the second order perturbation terms operated twice, \((J'' s_i \cdot s_j)^2\), with \( s_i \) the spin-1/2 operator discussed in Refs. 12 and 39, work in the similar manner. (b) Fluctuation between on-bond spin-1’s that are equivalent to those of panel (a). (c) In our spin-1 dimer system, the pair creation and annihilation term \((P)\) plays a major role which originates from the first order in \( J' \) and \( J'' \).
FIG. 9. (a), (b) Typical second order perturbation processes over three-dimers. (a) Processes of “correlated hopping” and (b) pair-creation of bosons. Ellipses mark the pair of sites to which the perturbation Hamiltonian $H_{\text{inter}}$ operates. (c) Energy levels of $H_{\text{intra}}$ of the three spin-1 dimer states. (d)–(g) $J'/J$ dependences of the ground state energies of the effective Hamiltonian $H_{\text{eff}}$ with and without $H_{3\text{body}}$ and the original spin Hamiltonian $H$ on the 9-dimer triangular lattice for $(B/J, (J' + J'')/J) = (d) (0.2, +0.2), (e) (0.2, -0.2), (f) (0.4, +0.2), \text{ and (g) (0.4, -0.2).}$