Spin nematics next to spin singlets

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We provide a route to generate nematic order in a spin-1/2 system. Unlike the well-known magnon-binding mechanism, our spin nematics requires neither the frustration effect nor a spin polarization in a high field or in the vicinity of a ferromagnet, but instead appears next to the spin singlet phase. We start from a state consisting of a quantum spin-1/2 singlet dimer placed on each site of a triangular lattice, and show that inter-dimer ring exchange interactions efficiently dope the SU(2) triplets that itinerate and interact, easily driving a stable singlet state to either Bose Einstein condensates or a triplet crystal, some hosting a spin nematic order. A variety of roles the ring exchange serves include the generation of a bilinear-biquadratic interaction between nearby triplets, which is responsible for the emergent nematic order separated from the singlet phase by a first order transition.

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Introduction.— Challenges in modern magnetism have been to clarify the role of the intrinsic quantum effect in exotic phases of matter. Spin-1/2 liquids\textsuperscript{12} that do not break any symmetry are characterized by the long range entanglement of their wave functions. Some phases break the symmetry quantum mechanically by forming the smallest entangled unit; a valence bond crystal is the long range order of a spin-1/2 singlet breaking translational symmetry\textsuperscript{13} and spin nematics is the SU(2)-symmetry-broken order of the quadrupole moment based on spin-1 pairs\textsuperscript{14}. The latter phase is our focus, and is established in a spin-1 bilinear-biquadratic Hamiltonian on square\textsuperscript{5,14} and triangular lattices\textsuperscript{6–10}. They appear whenever the biquadratic interaction $(S_i \cdot S_j)^2$, overwhelms the bilinear (Heisenberg) term where $S_i$ is the spin-1 operator. In fact, to entangle a pair of spin-1, one needs to exchange $(S_i^z, S_j^z) = (+1, -1)$ with $(-1, +1)$ by the biquadratic term, such that the spin-1/2 singlets are formed by a Heisenberg exchange, flipping $(+1/2, -1/2)$ to $(-1/2, +1/2)$. Unfortunately, the spin-1 biquadratic interaction is usually much weaker than the bilinear term which makes the nematic phase as elusive as spin liquids.

Moreover, spin-1 is not the basic magnetic unit in condensed matter, since it appears only as a triplet pair of spin-1/2’s of localized electrons. When the spin-1 is broken into pieces of 1/2, higher order exchanges among spin-1/2’s are required to realize the spin nematics. The simplest one is the four-body ring exchange interaction. When this interaction is applied to the polarized spin-1/2 magnets in a strong magnetic field or near the ferromagnetically ordered phases, a nematic order indeed appears\textsuperscript{12,13}, providing a good reference for a solid $^3$He\textsuperscript{14,17}. There, the ring exchange was ascribed a role to enhance quantum fluctuation among competing magnetic orders\textsuperscript{14,15}. Besides, this interaction generates a gapped spin liquid in a spin-1/2 triangular lattice antiferromagnet\textsuperscript{16,21}. The magnitude of ring exchange is enhanced near the Mott transition\textsuperscript{22} which may explain the origin of the spin liquid triangular lattice Mott insulator\textsuperscript{23} possibly realized in the organic $\kappa$-ET$_2$Cu$_2$(CN)$_3$\textsuperscript{24}. The ring exchange further supports the anomalous thermal magnon Hall transport in a kagome ferromagnet\textsuperscript{25}. Despite such rich physics relevant to the ring exchange, a clear-cut and systematic understanding of its role is still lacking.

This Rapid Communication shows how the ring exchange serves to yield a variety of quantum phases. We consider spin-1/2 antiferromagnetically coupled dimers forming a triangular lattice. When the ring exchange interactions are transformed into a bosonic language, they simultaneously play different roles: chemical potential, hoppings, and repulsive/attractive interactions. Particularly, the one called twisted ring exchange flips the $S_i$ pairs upside down, and contributes to the bilinear-biquadratic interactions. We obtain a rich phase diagram in the bulk limit that hosts Bose Einstein condens...
FIG. 2. (color online) Phase diagram of Eq. (1) on the plane of $K_R$ and $K_T$, with $J = 1$, $J'' = 0$ and $J = 2K_R$. The triangle and cross symbols indicate the first and second order transitions, respectively, obtained by analysis of exact diagonalization with $N = 12$ (24 spins). Solid lines are the analytical phase boundaries. By increasing $J'$ and $J''$, the nematic phase is stabilized toward smaller $K_R$ and $K_T$ (see Supplemental Material).

FIG. 3. (color online) (a) Density of triplets, $\langle n_t \rangle$ as a function of $K_R$. In BEC, $\langle n_t \rangle$ changes gradually. A nematic triangle and kagome have exactly $\langle n_t \rangle = 1$ and $3/4$, respectively. (b) Exact diagonalization energy per site $E/N$ of Eq. (1) as a function of triplet density, $n_t$, at $K_T = 0.05, 0.45$ and $K_R = 0 \sim 0.5$. (Upper panel) BEC phase with no anomaly. (Lower panel) Dips at $n_t = 0.3/4, 1$ indicate the three other stable phases in Fig. 2, and the first order transitions between them. (c) Structural factor of $\langle Q_i \cdot Q_j \rangle$ at the $K$ and $M$-points on the Brillouin zone boundary, together with the ones for $\langle S_i \cdot S_j \rangle$, characterizing 120° magnetic ordering (AFM). (d) Tower of states in the nematic triangular $\langle K_R = 0.1, K_T = 0.45 \rangle$ and BEC-AFM ($K_R = 0.5, K_T = 0$) phases.

sate (BEC) of $SU(2)$ bosons and nematic orders of triangular and kagome geometries.

**Model and Phase Diagram.** — Each lattice site consists of a pair of quantum spin-1/2 coupled by the antiferromagnetic Heisenberg interaction, $J$, as shown in Fig. 1(a). When we consider only $J(=1)$, the ground state is an exact product state of singlets with energy $E_0 = -NJ/4$. By introducing the inter-dimer interactions, our Hamiltonian is given as,

$$
\mathcal{H} = \sum_{i=1}^{N} JS_{i1} \cdot S_{i2} + \sum_{(i,j), \gamma=1,2} (J'S_{i\gamma} \cdot S_{j\gamma} + J''S_{i\gamma} \cdot S_{j\prime\gamma}) + \sum_{C} K_C \left( P_4 + P_4^{-1} \right) - \frac{9}{5} \sum_{(i\gamma,j\gamma) \in C} S_{i\gamma} \cdot S_{j\gamma} \quad (1)
$$

where $S_{i1}, S_{i2}$ are the spin-1/2 operators forming the $i$-th dimer, $J'$ and $J''$ are the Heisenberg (bilinear) exchanges, and $K_C$ denotes the four-body exchange (see Fig. 1(b)). Along the two different closed loops $C = R$ and $T$, the four spins permute both clockwise ($P_4$) and anticlockwise ($P_4^{-1}$), which we call the ring exchange (R) and twisted ring exchange (T) e.g. as shown in Fig. 1(c). The last two-body term appears when deriving the four-body interactions by the perturbation from the Hubbard model at half-filling [27,29].

Due to the ring exchange terms, a variety of phases emerges as shown in Fig. 2 when $K_T$ is small. $K_R$ drives the system to the BEC of triplets. At larger $K_T$, the nematic phases become dominant. Here, we stress that all the $S_i^z = 1, 0, -1$ component of triplets equivalently join this BEC, which is thus different from a magnon BEC (carrying a net magnetic moment) that typically appears in spin singlet systems by the magnetic field [19,33]. The spin nematics in a quantum spin-1/2 system known so far is based on a bound state of magnons created by the frustration effect, in a strong magnetic field or near the fully polarized ferromagnetic phase [13,14,15]. Our spin nematics does not require such frustration, and the overall feature of the phase diagram applies to square, honeycomb, and ladder systems as well [50].

**Bosonic representation.** — To understand the nature of the phase diagram, it is convenient to transform the basis in units of dimers rather than of spin-1/2 [37,38]. Each dimer hosts either a singlet ($S_i = S_{i1} + S_{i2} = 0$) or one of the triplets ($S_i = 1, S_i^t = 1, 0, -1$). Therefore, by regarding the singlet product state as a vacuum, we introduce a bosonic operator, $b_{i\alpha}^\dagger / b_{i\alpha}$ with $\alpha = 1, 0, -1$, which creates/annihilates a triplet with $S_i^{z} = 1, 0, -1$ on
an i-th dimer. Equation (1) is exactly transformed to $\mathcal{H} = \mathcal{H}_{tV} + \mathcal{H}_{\text{mag}} + \mathcal{H}_{\text{pair}}$ with

$$
\mathcal{H}_{tV} = \sum_{(i,j)\alpha} \left( t(b_{i\alpha}^\dagger b_{j\alpha} + b_{j\alpha}^\dagger b_{i\alpha}) + \nu_n n_j \right) - \sum_{i=1}^{N} \mu n_i
$$

$$
\mathcal{H}_{\text{mag}} = \sum_{(i,j)} \left( J (S_i^1 \cdot S_j^1) + B (S_i^1 \cdot S_j^2)^2 \right) n_i n_j
$$

$$
\mathcal{H}_{\text{pair}} = \sum_{(i,j)} P (b_{i1}^\dagger b_{j-1,1} + b_{i-1,1} b_{j,1} - b_{i1} b_{j,0} + b_{i0} b_{j1}) + \text{H.c., (2)}
$$

where $n_i = 0$ or 1 is the number of bosons under a hard core condition, and $S_i$ is the spin-1 operator on site-$i$ when it is occupied by a triplet, and fulfills $S_i^+ = b_{i1}^\dagger b_{i0} + b_{i0}^\dagger b_{i-1,1} - b_{i1} b_{i-1,1}$, $S_i^- = b_{i1}^\dagger b_{i0} + b_{i0}^\dagger b_{i-1,1}$, and $S_i^z = b_{i1}^\dagger b_{i1} - b_{i1} b_{i1}$. The magnetic interactions, $J$ and $B$, will be discussed shortly. The parameters are given as,

$$
\mu = -J + \left( \frac{96}{5} K_R - \frac{24}{5} K_T \right) \nu_{\alpha} (3)
$$

$$
t = \frac{1}{2} (J' - J'' + K_R) \nu_{\alpha} (4)
$$

$$
V = 4 (K_R - K_T) \nu_{\alpha} (5)
$$

$$
P = \frac{1}{2} (J' - J'') - K_R \nu_{\alpha} (6)
$$

In the present Rapid Communication, we take $J' = 2K_R$ and $J'' = 0$, in order to keep $P = 0$, which allows for the exact evaluation of the phase boundaries, and the number of triplets is conserved. The role of $P \neq 0$ is mainly to enhance $B$ and stabilize the nematic order (see Supplemental Material), while the physics itself is not influenced qualitatively.

As the chemical potential, $\mu$, gets lower with $K_R$, the bosons are doped to the vacuum (singlet). Meanwhile, there arises an interaction between doped bosons. Figure 3(a) shows how the occupation of the triplet density, $n_t \equiv \sum_i n_i / N$, develops, where $n_i$ is the value that gives the minimum of total energy $E$ among all different $n_t$-sectors in Fig 3(b). At $K_R > K_T$, the kinetic energy gain due to $t$ favors dopping the bosons, but the repulsive interaction $V > 0$ does not, thus $\langle n_t \rangle$ increases gradually due to their competition. Contrarily at $K_R < K_T$, the attractive $V < 0$ helps $\mu$ to dope triplets and there occurs a first order transition from the $\langle n_t \rangle = 0$ to the $\langle n_t \rangle = 1$ phase, which is also visible in the dip of energies at $n_t = 0$ and 1 in Fig 3(b). Once all the sites are occupied by triplets, their spin-1’s interact via $\mathcal{H}_{\text{mag}}$, which takes a well known form called the bilinear-biquadratic interaction, with $B = 2K_R$ and $J = (-K_R + 4K_T) / 5 + (J' + J'') / 2$. In a triangular lattice, the spin-1 bilinear-biquadratic Hamiltonian is known to host a nematic long range order when $B > J$ and the equivalent condition, $K_T > 3K_R/2$, is actually fulfilled in our nematic triangular phase.

Spin nematic order. — The quadrupolar moment is the order parameter of the spin nematics and is described by a symmetric and traceless rank-2 tensor, $Q_{ij}^{\alpha\beta} = S_i^\alpha S_j^\beta + S_j^\alpha S_i^\beta – 2S^\alpha S^\beta / N$. We examined its two point correlation whose structural factor takes a peak at the $K$- and $M$-points at the Brillouin zone boundary, which are plotted as functions of $K_R$ in Fig 3(c). A dominant peak at the $K$-point is consistent with the previously reported antiferro-quadrupolar ordering (AFQ) on the triangular lattice. The peak of the spin-spin correlation function at the $K$-point, characterizing the 120° antiferromagnetic (AFM) ordering is suppressed in these regions.

To further confirm the existence of nematic long range order, we show the energy spectrum at $K_T = 0.5$ and $K_R = 0.1$ in Fig 3(d). There actually appears a tower of low-lying energy levels well separated from the other excitations, and the symmetries of the quasidegenerate joint states (QDJS) belonging to different spin sectors follow those already known for the SU(2)-symmetry-broken spin nematics on a triangular lattice.

Twisted ring exchange. — We need to understand why the biquadratic $B$ in $\mathcal{H}_{\text{mag}}$ (Eq. 2) originates from the twisted ring exchange, $K_T$, and not from $K_R$. The spin-1 biquadratic term exchanges the up and down spin-1 pairs, changing $S_i^z$ by ±2, while the Heisenberg (bilinear) term flips $S_i^z$ only by the ±1. Figure 4(d) shows that when $K_R$ is operated to the four spin-1/2’s on a plaquette, they move cyclically, and transform the dimer spin $(S_i^z, S_j^z) = (+1, -1)$ to $(0, 0)$. Whereas, if the path is twisted, $K_T$ can move the two spin 1/2’s on one dimer to the other dimer at once, and flip the dimer spin $(+1, -1)$ to $(-1, +1)$, contributing to the biquadratic term.

The magnitude of $K_T$ had been considered as small in a quantum spin system, as it originates from the fourth order perturbation in a Mott insulator. According to our evaluation, the on-site Coulomb interaction $U$ against the inter-dimer transfer integral, $t_{ij}$, should be
$U/t_{ij} \lesssim 7$ in order to have $K_C/J \gtrsim 0.1$, which is not too unrealistic. It is also shown that in the vicinity of the Mott transition, $U/t \sim 8$, the ring exchanges can be as large as $J'/J$. We further mention that $P \neq 0$ works as an effective biquadratic term, thus a larger $J' - J''$ will stabilize the nematic phase than the one found in Fig. 3 (see Supplemental Material).

Instabilities.— The phase boundaries of Fig. 2 can be determined half-analytically by examining the energetics of the hard core bosonic model, $H_{AV}$, in the bulk limit. The phase diagram of $H_{AV}$ for $t > 0$ is shown in Fig. 3(a). Similar to the case of $t < 0$ studied in the context of cold atoms, the $1/3$- and $2/3$-filled crystal phases appear at large $V/t$, and the supersolid phases in between. The other parts are divided into the vacuum, bosonic BEC, and a solid, and their boundaries are exactly determined. The onset of the BEC from the vacuum is given by the kinetic energy gain of a single boson, $\mu = -3t/2$. The first order transition line between the vacuum and the solid takes place at $\mu = 3V$, as the attractive interaction favors all the triplets to be doped at once by maximally gaining the energy $3V < 0$. Finally, the instability of the solid against the BEC is evaluated by the energy of a doped hole, $-3t - 6V$.

The shaded region in Fig. 4 covers the parameter range of Fig. 2. The onset of BEC mapped to our model is, $K_{AV} > K_T$ in good agreement with the one from the exact diagonalization. In regions with higher boson densities, the bosons interact magnetically, thus we need to take account of the effect of $H_{\text{mag}}$ terms on the energy of hard core bosons. For this purpose, we introduce an effective interaction including the corrections from the magnetic terms, $V_{\text{eff}} = V + \langle J(S_i \cdot S_j) + B(S_i \cdot S_j)^2 \rangle$, and evaluate its value by separately analyzing the spin-1 bilinear-biquadratic Heisenberg model. Originally, the upper left-half of the phase diagram, $K_T > K_R$ was the region with an attractive interaction $V < 0$. However, this correction pushes the phase boundary upward, and the repulsive $V_{\text{eff}} > 0$ region starts just below the triangular nematic phase. The boundary between the singlet and the nematic triangle is given by $\mu = 3V_{\text{eff}}$.

Nematic kagome phase.— There is another phase in the middle of the diagram with $\langle n_t \rangle = 3/4$, characterized by the peak of the AFQ order at three $M$-points (see Fig. 3(c),(d)). The spatial structure of this quadrupole order expected is a kagome geometry that is realized by regularly depleting one quarter of the lattice sites of the triangular lattice. However, in a pure hard core bosonic model, there is no reason to favor such a kagome structure which is indeed absent in Fig. 3. We thus reexamine the energy of the original Hamiltonian for all different $n_t$ sectors in Fig. 3(b); there is a dip in the energy at $\langle n_t \rangle = 3/4$, which competes with $\langle n_t \rangle = 1$ when $K_T > 0.340$. This is in sharp contrast to the smooth $n_t$ dependence of $E$ in the BEC region. The competition between the three discrete fillings gives the first order transitions. The energy dip at $\langle n_t \rangle = 3/4$ comes solely from the magnetic interaction, $\langle H_{\text{mag}} \rangle$, and not from $\langle H_{\text{AV}} \rangle$ (Supplemental Material Fig. S2), to be more precise, by the contribution from $B(S_i \cdot S_j) = B/2(\langle Q_i \cdot Q_j \rangle - \langle S_i \cdot S_j \rangle) + \text{const}$. This also suggests that the bosons remain BEC. We calculate the bond energy of the biquadratic Hamiltonian on a kagome lattice, and find that it is lower by $dE_{\text{mag}} \sim 0.05$ compared to the same Hamiltonian on the triangular lattice. Thus, the phase boundary in Fig. 2 is finally corrected to, $\mu = 3t + dE_{\text{mag}} + 6V_{\text{eff}}$, showing excellent agreement with the ones obtained by the exact diagonalization.

Remarks.— The mechanism to generate a variety of phases in the spin-1/2 dimer model by the ring exchange interactions is fully fixed, by the exact transformation to a hard core bosonic language. We would like to stress the following points: The prototypes of emergent BEC’s in spin systems were to dope magnons to a spin singlet state by the magnetic field, whereas, here, the ring exchange interaction serves as a fictitious field that does not break the SU(2) symmetry, and dopes the SU(2) bosons, not the magnons carrying magnetization. Once the bosons are doped, the bilinear-biquadratic interaction induced by a four-spin-exchange along the twisted path works to stabilize the nematic orders. The spin nematics basically requires an exchange of spin-1 bosons, thus was observed in the spin-1/2 system in a strong magnetic field (spin polarized state) or in the vicinity of frustrated magnetism, where the frustration played a key role to enhance the quantum fluctuation. Our spin singlet state is a trivial product state in the contour extreme limit of such a complication, thus one may find it rather counterintuitive to have a nematic phase next to it.

The spin-1 hard core bosonic model can also be regarded as a strong coupling limit of the spinor boson systems studied in cold atoms. There, the bosons are softly exclusive on each site due to the on-site interaction, $U$ and the second order perturbation from the $U/t \to \infty$ limit gives a biquadratic interaction between spin-1 bosons, and the nematic Mott insulating phase appears. A situation similar to man-made optical lattices is naturally realized in our quantum spin-1/2 model representing crystalline solids, as such a dimer system is actually quite ubiquitous in transition metals such as BaCuSi$_2$O$_6$, Ba$_2$CoSi$_2$O$_6$Cl$_2$, and Ba$_3$MRu$_2$O$_9$. In Ba$_3$MRu$_2$O$_9$, a nonmagnetic phase is actually found next to the singlet phase, and the relevance with our findings remains an issue to be clarified.

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There is another term $K_{K'}$, comparable to $K_t$ and $K_T$ as shown in Ref.\textsuperscript{22}, but plays an equivalent role with $K_t$, thus is abbreviated in the present paper.

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