Spin nematics is a phase of matter without magnetic order, but still breaks the spin-rotation symmetry \([\mathbb{I}]^2\). This exotic state established itself as an intermediate category of magnetism and quantum spin liquids, with an advantage in that it could be captured much more easily than the spin liquids by a signature of symmetry breaking, both in material systems and in toy lattice models. A more general description of spin nematics is “a quadrupole order of quantum order perturbation theory. It turns out that the biquadratic exchange interaction between spin-1, which is one of the origins of the nematic order, arises only in processes where the four different electrons exchange cyclically along the twisted loop, which we call “twisted ring exchange” processes. We show that the term becomes the same order with the Heisenberg exchange interactions when the on-orbital Coulomb interaction is not too large. Whereas, the inter-orbital Coulomb interactions give rise to additional processes that cancel the twisted ring exchange, and strongly suppresses the biquadratic term. The Mott insulator with two electrons on degenerate two orbitals is thus not an ideal platform to study such nematic orders.

![FIG. 1. Mott insulators hosting one electron per orbital, where the orbital-1 and 2, orbital-3 and 4 form pair-A and B, respectively. Case (i) considers each pair of orbitals as “dimer” and Case (ii) as ion (site) having degenerate orbitals which is realized in the \(e\)-electron systems. Case (ii’) discussed in Ref. \([4]\) takes account of an additional quasi-degenerate energy level, where the loop similar to process-T between the excited states yields a biquadratic term of the same order with a bilinear term.](image-url)
The Hamiltonian contains the so-called ring exchange term, \( H_\text{eff} = B \sum_i \langle n_i \rangle \), with spin \( \sigma \), where \( n_i = c_{i\sigma}^\dagger c_{i\sigma} \) is the number operator. We consider three different species of transfer integrals, \( t \), \( t' \) and \( t'' \) shown in Fig. 1. When \( U \) is strong enough, each orbital is occupied by a single electron, and the low energy degrees of freedom is the spin-1/2 on each orbital, \( s_i = \pm 1/2 \). The effective Hamiltonian at second order perturbation at \( t_j/U < 1 \) is the well-known Heisenberg term, \( H_\text{eff} = \sum_{\langle i,j \rangle} J_{ij} s_i \cdot s_j \), with \( J_{ij} = 4t_j^2/U \). The third order terms cancel out on the whole.

The fourth order terms are classified into four categories shown in Figs. 2(a) and 2(b) as \( H_\text{eff} = H_\text{eff}^{(1b2s)} + H_\text{eff}^{(2b3s)} + H_\text{eff}^{(2b4s)} + H_\text{eff}^{(4b4s)} \), hopping processes taking place along one, two, and four different bonds (1b, 2b, 4b) and over two to four different sites (2s, 3s, 4s), are denoted as (1b2s), (2b3s), (2b4s), and (4b4s), respectively. The (1b2s) and (2b3s) contribute to the Heisenberg interaction, and all the (2b4s) cancel out. The (4b4s) consisting of hoppings along four all different bonds yields the so-called ring exchange term,

\[
H_\text{eff}^{(4b4s)} = -\frac{4K_C}{5} \sum_{\langle i,j \rangle, a,b,c,d} s_i \cdot s_j + 4K_C \sum_{a,b,c,d} \left[(s_a \cdot s_b)(s_c \cdot s_d) + (s_a \cdot s_d)(s_b \cdot s_c) - (s_a \cdot s_c)(s_b \cdot s_d)\right],
\]

where we take the hoppings along the closed loop \( C \), consisting of \( a-b-c-d-a \). As shown in Fig. 2(b), there are three different closed paths of fourth order that contribute to \( H_\text{eff}^{(4b4s)} \); the first one using \( t \) and \( t'' \) along \( (a,b,c,d) = (1,2,4,3) \) (\( C = R \)), which we call process-R, is a typical ring exchange first discussed by Takahashi. The one along \( (1,2,3,4) \) is denoted as process-R', and the last one \( (1,3,2,4) \) as process-T. The contributions to the effective Hamiltonian from these processes are given by assigning the indices of spins \( a-d \) in Eq. (3) the orbital indices 1-4 along the closed paths of \( C = R, R', \) and T. The processes-R' and T are derived already by Calzado and Malrieu as extra four-body-spin exchange processes. Equation (3) differs by a factor in the first term from the ring exchange in solid \( ^3 \text{He} \) [18, 19], which is described by \( (P_4 + P_4') \) with an operator \( P_4 \) permutating the spins counterclockwise.

The next step is to transform the above expression of \( H_\text{eff}^{(n)} \) \( (n = 2,4) \) by the spin-1/2 degrees of freedom defined on each orbital into the spin-1 degrees of freedom on each pair of orbitals. This is done by projecting the effective Hamiltonian to a basis of triplets using the projection operator, \( P_3 \), as \( H_\text{eff}^{(n)} = P_3 \sum_{\text{process}} B_4 \), where we find an effective Hamiltonian of the form of Eq. (1) with the coupling constants given separately for each process as

\[
J = J_2 + \sum_{\text{process}} J_4^{\text{(process)}}, \quad B = \sum_{\text{process}} B_4^{\text{(process)}},
\]

where the subscript indicates the order of perturbation, and the “process” indicates (1b2s), (2b3s), etc. For the Heisenberg interaction...

**FIG. 2.** (a) Fourth order processes over less than three bonds. (b) Three different (4b4s) processes consisting of closed loops, which refer to the typical ring exchanges (R, R') and twisted ring exchange (T). The processes require all the four hopping terms along these loops to be present. (c) One of the processes that contribute to the spin-1 biquadratic exchange, where \( \uparrow \downarrow \) flips to \( \downarrow \uparrow \). (d) Typical disconnected processes that cancel out and do not contribute to \( B \) at \( U' = 0 \). (e) Evaluation of model parameters, \( J \) and \( B \), of Eq. (4), in Case (i) based on spin-1 degrees of freedom per site, with \( U' = 0, 1, 2 \) and \( t = t' = t'' = 1 \) as a function of \( t/U \). The contributions at \( U' \neq 0 \) in Eqs. (12)–(15) are included.
terms, we find

\[ J_2 = 2(t^2 + r^2)/U, \]

\[ J_4^{(1b2s+2b3s)} = (-8t^4 - 8t^2r^2 + 4r^2t^2 + 4t^2r^2)/U^3, \]

\[ J_4^\text{(R)} = -K_r/5 = -4r^2t^2/U^3, \]

\[ J_4^\text{(T)} = -K_r/5 = -4r^2t^2/U^3, \]

\[ J_4^\text{(T)} = 4K_T/5 = 16r^2t^2/U^3. \]

At this stage, we see that the contributions from (4b4s) severely depend on the geometry of paths, and in fact, the biquadratic term of spin-1 appears only in process-T as

\[ B_4^\text{(T)} = 2K_T = 40r^2t^2/U^3. \]  

This could be understood more intuitively as follows; let us explicitly show a matrix representation of the biquadratic term of spin-1, respectively. One of the main roles of the spin-1 biquadratic exchange term is to flip the pairs as, \(|S_A^z + S_B^z = 0\rangle\) space as

\[ \langle ml | (S_A^z \cdot S_B^z)^2 | m'l'\rangle = \begin{pmatrix} 2 & 1 & 1 & 1 \\ -1 & 2 & -1 & -1 \\ 1 & -1 & 2 & -1 \\ 1 & -1 & -1 & 2 \end{pmatrix}. \]

where the three basis states are chosen as \(|m\rangle = |_{\downarrow \downarrow \uparrow \uparrow\rangle}, |_{\uparrow \downarrow \uparrow \downarrow\rangle}, |_{\downarrow \uparrow \uparrow \downarrow\rangle}\), with \(\downarrow, \uparrow\) indicating \(S^z = 1, 0, -1\) of each spin-1, respectively. One of the main roles of the spin-1 biquadratic exchange term is to flip the pairs as, \(|_{\downarrow \downarrow \downarrow \downarrow\rangle}\), and vice versa (\((1, 3)\) and \((3, 1)\) elements of Eq. (11)). Here, decomposing these \(S = 1\) and \(S^z = \pm 1\) spins into the triplets of \(\pm 1/2\) on orbitals \(1\) and \(2\), while \(|_{\uparrow \downarrow \uparrow \downarrow\rangle} = |_{\downarrow \uparrow \downarrow \uparrow\rangle}\), we recall the perturbation processes. As shown in Fig. 3(c), part of the process-T contributes to this spin flip. If we try to do the same thing in process-R and R', it immediately breaks down. This is because in order to make this flip, we need to transfer the spin electrons on orbital-1 and 2 to orbital-3 and 4, and the down spin electrons on orbital-3 and 4 to orbital-1 and 2, while to do so via ring exchange process, orbital-1 needs to be connected with both orbital-3 and 4 and so as orbital-2 along the loop. One may think that the (2b4s) processes, hopping back and forth along the two different bonds, may also flip the spins in the above mentioned manner. However, such processes are basically a combination of two independent second order exchange processes, which we call “disconnected processes”, and cancel out on the whole (see Fig. 3(d)). This is natural because, otherwise, arbitrary choices of two independent bonds in the bulk system will generate numbers of magnetic long range interactions no matter how distant they were separated. Thus, we finally find that the twisted ring exchange (T) is responsible for the biquadratic interaction, whereas the ordinary ring exchange (R and R') do not. This is the main message to deliver in the present paper. As shown in Fig. 3(e), the evaluated B and J in Eq. (2) (20), take the same order when \(U \lesssim 5t\), which is not too unrealistic.

Notice that, although we projected out the \(S = 0\) (singlet) state of each site, there are finite terms between singlets and triplets. Namely, \(P_1H_{\text{eff}}^{(0)}(1 - P_1)\) and \((1 - P_1)H_{\text{eff}}^{(0)}(1 - P_1)\) are not at all negligible both at \(n = 2\) and 4. However, these terms work to control the population of triplets and singlets, which will be discussed elsewhere [21], and thus only indirectly contribute to the magnetic properties as they do not yield any magnetic exchange interaction.

One extension of Case (i) is to add the intra-dimer Coulomb interactions, \(H_d = U'(n_1n_2 + n_3n_4)\), to Eq. (2). After performing the same perturbation calculation, one finds that Eqs. (5)–(8) do not change. However, there emerges an additional contribution from the disconnected (2b4s) processes as

\[ B_4^{(2b4s; U')}(U') = \frac{4t^2 + r^2}{U^2} \left( \frac{2}{U} - \frac{1}{U - U'} - \frac{1}{U + U'} \right). \]

This is simply because, at finite \(U'\), the two bonds are no longer disconnected. The ones from process-T are corrected from Eqs. (5) and (10), and \(J_4^{(2b4s; U')}(U')\) is added as

\[ J_4^{(2b4s; U')}(U') = 16r^2t^2/U^2(U - U'), \]

\[ B_4^{(2b4s; U')}(U') = t^2r^2 \left[ 32/3(U - U') + 8/3(U + U') \right], \]

\[ J_4^{(2b4s; U')}(U') = 16r^2t^2 \left[ -1/U^3 + 1/U^2(U - U') \right]. \]

By further adding the Hund interaction between the dimerized two orbitals, we find Case (ii). We consider a Kanamori Hamiltonian \([22, 23] H = H_0 + H_d,\) with

\[ H_d = \sum_{\sigma; ab \neq \pm} \sum_{\sigma} n_{a\sigma}n_{b\sigma} + U' \sum_{\sigma} (n_{a\uparrow}n_{b\downarrow} + n_{a\downarrow}n_{b\uparrow}) + J_H \left( c_{a\uparrow}^\dagger c_{a\uparrow} c_{b\downarrow}^\dagger c_{b\downarrow} + c_{a\downarrow}^\dagger c_{a\downarrow} c_{b\uparrow}^\dagger c_{b\uparrow} \right) + J_F \left( c_{a\downarrow}^\dagger c_{a\downarrow} c_{b\uparrow}^\dagger c_{b\uparrow} + c_{a\uparrow}^\dagger c_{a\uparrow} c_{b\downarrow}^\dagger c_{b\downarrow} \right), \]

where the orbital indices \((a, b) = (1, 2), (3, 4)\) are those on the same site. The inter-orbital intra-site Coulomb interaction \(U'\) and the Hund coupling \(J_H\) are taken as such that they fulfill \(U = U' + 2J_H\), in crystal fields of cubic symmetry. Therefore, Case (ii) roughly corresponds to the large-\(U'\) version of Case (i). The \(J_F\)-term expresses the pair hopping. As the two degenerate pairs of orbitals are orthogonal, we set \(t^2 = 0\), which has the geometry of the twisted ring exchange.

The perturbation process is rather complicated as both \(J_H\) and \(J_F\) hybridize the electronic states belonging to the same site when there are two electrons. The representative two-electron eigenstates of \(H_0 + H_d\) (while taking \(t_{ij} = 0\)) are given in Fig. 3(a). In the following, we call the state which has \(n_A\) and \(n_B\) electrons on site-A and B as \((n_A + n_B)\)-electron state. In Case (i), the low energy manifold was confined to those with one electron per orbital, but here, since \(U'\) differs from \(U\) only by \(2J_H\), the one with the doubly occupied single orbital \(|\uparrow\downarrow\rangle\) on either of the two sites is also included in the low energy manifold, which we denote \(|m\rangle\). Therefore, \(|m\rangle\) consists of all \((2+2)\)-electron states, and the excited states, \(|l\rangle\), are the \((1+3), (3+1), (0+4), (4+0)\)-electron states (see Fig. 3(a)). Besides taking account of hybridization of states, we also need to treat the processes differently from Case (i) in classifying them into two groups; \(|m\rangle - |l\rangle - |\bar{l}\rangle - |\bar{m}\rangle\) has \(|m\rangle\) and \(|l\rangle\) in the middle, and \(|m\rangle - |l\rangle - |\bar{m}\rangle - |\bar{l}\rangle\) is in the middle (see Fig. 3(b)). The Hund’s coupling generates several paths to the second and third hopping processes, as it allows
electron states, we project them onto the states with $H$ on each site consisting of one electron per orbital via $P$ found to suppress in overall both $J_1$ and $J_2$ of the same energies as listed. The right half of the (2+2)-ele states have the double occupancy of orbitals. There are many other configurations not shown, having the same energies as listed. The right half of the (2+2)-ele states have the double occupancy of orbitals. (b) Twisted ring exchange processes modified from those of Fig. 2(c) when $J_p \neq 0$. (c) Evaluation of model parameters of Case (ii), with $t = t' = 1$, $J_p = 0$, and $U = 6$.

The fourth order biquadratic terms are evaluated separately for each process [24] and the dominant contribution comes from the twisted ring exchange (T) process and the (2b4s) ones, which are given for the case of $J_p = 0$ as

$$B_4^{(T)} = \frac{t^4}{2} \left( \frac{12}{a^2 e} - \frac{4}{a d} \right),$$

and

$$B_4^{(2b4s)} = \frac{t^4}{2} \left( \frac{4}{a^2 d} + \frac{4}{a d^2} + \frac{2}{a b} + \frac{2}{a b^2} \right).$$

with $a = U + J_H$, $b = U - J_H$, $c = U - 3J_H$, $d = U - 5J_H$, $e = U - 4J_H$, $f = U - 2J_H$. (The contribution from $J_p \neq 0$ is not large [24]). Figure 3(c) numerically evaluates $J$ and $B$ including all the processes up to fourth order [24]. For the Heisenberg terms, the contribution from the fourth order, $J_4$, is ferromagnetic and suppresses the antiferromagnetic $J_2$. Regarding the biquadratic term, the negative contributions from $B_4^{(T)}$ is suppressed by the positive contributions from $B_4^{(2b4s)}$, and resultantly, the value of $B$ becomes small by one orders of magnitude, compared to $J_4$. We finally notice that the $J_H \to 0$ limit of our results in Case (ii) is not connected to $U' \to U$ of Case (i). This is because the formulation of Case (i) is valid at $U' \ll U$ and the $U = U' + 2J_H \sim U$ region is properly described only in Case (ii). The calculation taking the doubly occupied (2+2)-states as $|l\rangle$ is discussed in Ref. [25].

From the comparison of Case (i) and (ii), we find that the values of $J$ do not differ much, whereas $B$ takes the value comparable to $J$ only in the former case, as $J_H$ is usually much smaller than $U$ and $U'$. Therefore, the spin-1 system based on the two-orbital Mott insulator represented by Case (ii) does not afford sufficient degree of biquadratic interaction. The idea to overcome this issue is given by Mila and Zhang [14], who took account of one extra orbital to each site in Case (ii) that is quasi-degenerate but higher in energy by $\Delta$, which is shown in Fig. 1 as Case (ii’). They found that second order processes of particles hoppings to the third orbitals on the neighboring sites and coming back will give negative sign to the Heisenberg term, and will cancel out the Heisenberg term at that order on the whole. There are closed T-shaped paths consisting of two $t_{ex}$’s and $t'$’s, connecting one of the degenerate orbitals and one excited states on each site. The fourth order process along this T-shaped path gives rise to additional biquadratic term, which becomes the same order as $J$ when $t_{ex} \sim t'$.

We finally discuss the possible relevance of process-T and the bulk nematic order. For the spin-1 to be relevant in a system based on Case (i), we have to need the effective ferromagnetic coupling within each dimer, namely between orbital-1 and 2, and between orbital-3 and 4. Placing a strong magnetic field is known to be effective [13]. One way to realize such ferromagnetic dimers is to have an indirect hopping between dimerized orbitals mediated by the extra orbitals placed off the dimer bonds, and the Kanamori–Goodenough rule allows the exchanges to become ferromagnetic [25]. This picture is close to the above mentioned protocol by Mila and Zhang [14].

Another way is to consider a toy model, a uniform square lattice with ferromagnetic nearest neighbor exchange interactions, and antiferromagnetic next-nearest neighbor ones in the diagonal direction. Indeed, this kind of construction is
called $J_1$-$J_2$ square lattice model, and is known to yield a nematic order $[12]$. While the origin of such nematic order is attributed to frustration, we find that the next-nearest neighbor exchange interactions work together with the neighboring ferromagnetic exchanges and form process-$T$, yielding the effective biquadratic term, which shall be a microscopic explanation of what is known so far in numerics.

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1 Interaction coefficients for Case (i)

While only the sums of the parameters are shown in the main text, we compare each interaction parameter appeared in Case (i) below (see Fig. S1).

![Graph showing interaction coefficients for Case (i)](image)

Fig. S1: Evaluation of model parameters in Case (i) as a function of $U/t$. We set $U' = 1$ and $t = t' = t'' = 1$.

2 Interaction coefficients from 4th order perturbation for Case (ii)

Here we note the interaction coefficients of the bilinear (Heisenberg) interaction $\hat{S}_i \cdot \hat{S}_j$, and the biquadratic interaction $\left(\hat{S}_i \cdot \hat{S}_j\right)^2$, for 2-site-2 orbital system (Case (ii)). We include $J_p \neq 0$ in the following equations, although only part of the terms at $J_p = 0$ is given in the main text.

**Bilinear (Heisenberg) interactions**

Each element of $J_4^{(\text{process})}$ is expressed as follows:

\[
J_4^{(T)} = t^2t'^2\left[-\frac{12}{(U + J_H)^2(U - J_H - J_p)} - \frac{4}{(U - 2J_H - J_p)^2(U + J_H)}\right],
\]

\[
J_4^{(2b4s)} = t^4 + t'^4\left[-\frac{8}{(U + J_H)^3} + \frac{6}{(U + J_H)^2(U - J_H)} + \frac{2}{(U + J_H)(U - J_H)^2}\right],
\]

\[
J_4^{(1b2s)} = t^4 + t'^4\left[-\frac{8}{(U + J_H)^3} - \frac{6}{(U + J_H)^2(U - J_H)} - \frac{2}{(U + J_H)(U - J_H)^2}\right],
\]

\[
J_4^{(2b3s)} = t^2t'^2\left[-\frac{16}{(U + J_H)^3} - \frac{12}{(U + J_H)^2(U - 2J_H - J_p)} - \frac{4}{(U + J_H)(U - 2J_H - J_p)^2}\right].
\]
Biquadratic interactions

Each element of \( B_4^{\text{process}} \) is expressed as follows (\( B_4^{(T)} \) and \( B_4^{(2b4s)} \) are appeared also in the main text):

\[
B_4^{(T)} = r^4 \left[ \frac{12}{(U + J_H)^2(U - 4J_H - J_p)} - \frac{4}{(U - 4J_H - J_p)^2(U + J_H)} + \frac{4}{(U + J_H)^2(U - J_H)} \right],
\]

\[
B_4^{(2b4s)} = \frac{r^4 + r'^4}{2} \left[ \frac{4}{(U + J_H)^3} + \frac{6}{(U + J_H)^2(U - J_H)} - \frac{2}{(U + J_H)(U - J_H)^2} + \frac{2}{(U + J_H)^2(U - 3J_H)} + \frac{1}{(U + J_H)(U - 3J_H)^2} \right],
\]

\[
B_4^{(1b2s)} = \frac{r^4 + r'^4}{2} \left[ \frac{4}{(U + J_H)^3} - \frac{6}{(U + J_H)^2(U - J_H)} + \frac{2}{(U + J_H)(U - J_H)^2} + \frac{3}{(U + J_H)^2(U - 3J_H)} + \frac{1}{(U + J_H)(U - 3J_H)^2} \right],
\]

\[
B_4^{(2b3s)} = \frac{r^4 + r'^4}{2} \left[ \frac{8}{(U + J_H)^3} - \frac{6}{(U + J_H)^2(U - 3J_H)} - \frac{2}{(U + J_H)^2(U - 3J_H)^2} - \frac{12}{(U + J_H)^2(U - 2J_H - J_p)} + \frac{4}{(U + J_H)(U - 2J_H - J_p)^2} + \frac{4}{(U + J_H)(U - 4J_H - J_p)^2} \right].
\]

Figure S2 shows the comparison of the interaction parameters in Eqs. (S1)–(S8) at \( J_p = 0 \).

Fig. S2: Evaluation of model parameters in Eqs. (S1)–(S8) as a function of \( J_H/t \) at \( U = 6 \) and \( J_p = 0 \). We set \( t = t' = 1 \).