Spin-Nematic Vortex States in Cold Atoms

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The (pseudo-)spin degrees of freedom greatly enriches the physics of cold atoms. This is particularly so for systems with high spins (i.e., spin quantum number larger than 1/2). For example, one can construct not only the rank-1 spin vector, but also the rank-2 spin tensor in high spin systems. Here we propose a simple scheme to couple the spin tensor and the center-of-mass orbital angular momentum in a spin-1 cold atom system, and show that this leads to a new quantum phase of the matter: the spin-nematic vortex state that features vorticity in an SU(2) spin-nematic tensor subspace. Under proper conditions, such states are characterized by quantized topological numbers. Our work opens up new avenues of research in topological quantum matter with high spins.

Introduction — Cold atoms are well known to provide an ideal platform for quantum simulation [1]. As a quantum simulator, cold atoms can not only simulate important toy models arising from other subfields of physics, but also offer opportunities to construct new models that take advantages of their unique properties. One particular example is synthetic spin-orbit coupling (SOC) generated either by Raman laser coupling [2–5] or by periodic modulation [6–8], due to the flexibility of tailoring laser configuration or Floquet engineering, novel types of SOC not naturally occur in other systems can be realized. Another unique property of the atom is that the number of internal states, (i.e., the spin) involved can be tuned to some extent, which makes possible the exploration of intriguing physics of high spins [9].

Combining SOC and high spin, SOC in cold atoms with high spins has received much attention in recent years. Raman laser induced SOC in spin-1 condensate [10] was realized in the group of Spielman [11], where various phase transitions and the associated quantum tricritical point have been identified. Very recently, interesting phenomena have been explored in a novel type of coupling between the center-of-mass orbital angular momentum (OAM) and the spin vector in spin-1 condensates [12–14] where topological spin vortices, as well as the Hess-Fairback effect have been observed.

Nevertheless, previous works, including the studies mentioned above, predominantly focus on the textures of the spin operators but few on those of the nematic tensors [9, 15, 16]. The nematicities, which serve as fundamental quantities in high-spin quantum systems, have proved to be of wide usage in distinguishing different phases [9, 16] or generating topological structures [9, 17, 18]. In the current work, we propose to synthesize the coupling between the OAM and the spin-nematic tensor in a spin-1 condensate, and show that this coupling leads to a novel vortex state in a special SU(2) subspace spanned by a combination of spin and nematic operators. We call such states, which have never been studied before, the spin-nematic vortex state.

Spin nematic and OAM coupling — Our proposal builds upon previous studies on coupling spin and OAM [12] and on coupling spin tensor and linear momentum [19] in cold atoms. Specifically, we consider a spin-1 condensate harmonically confined in a pancake-shaped trap, where the axial confinement along the z-axis is much stronger than that in the transverse directions. As schematically shown in Fig. 1, the three spin states with magnetic quantum number $m_F = 1, 0$ and $-1$ are Raman coupled by three co-propagating Laguerre-Gaussian beams along the z-axis. Two of the laser beams carry OAM $L_z = \hbar$ and the third beam has $L_z = -\hbar$. In this configuration, all the essential physics occurs in the transverse plane and hence we can model the system as a quasi-two-dimensional one. In the lab frame, the single-particle Hamiltonian can be written in the polar coordinates $(r, \phi)$ as (taking $\hbar = m = \omega = 1$ where $m$ is the atomic mass, and $\omega$ the transverse trap frequency) [20]:

$$H_0 = -\frac{1}{2} \nabla^2 + \frac{1}{2} r^2 + \delta S_z + q S_z^2$$

$$+ i \sqrt{2} \Omega_R (r) \left( e^{i \phi} |z\rangle \langle y| - e^{-i \phi} |y\rangle \langle z| \right),$$

where $S = \{S_x, S_y, S_z\}$ are the spin operators, $\delta$ the two-photon Raman detuning, $q$ the effective quadratic...
Zeeman energy, $\Omega_R(r) = 2\Omega_0 \left( \frac{\mu}{2} \right)^2 e^{-2r^2/w^2}$ the $r$-dependent Raman coupling strength with $\Omega_0$ characterizing the Raman beam intensity, and $w$ the beam width. We have neglected the ac-Stark shift from the Raman beams. This can be achieved by using the tune-out frequency as were done in recent experiments [13, 21, 22]. In Eq. (1), instead of the bare spin states $|m_F = +1, 0\rangle$, and $| - 1, 0\rangle$, we have used the Cartesian polar states $|\mu\rangle$ with $\mu = x$, $y$ and $z$, which are defined as the eigenstates to spin operator $S_\mu$ with zero eigenvalue, i.e., $S_\mu |\mu\rangle = 0$. In terms of the bare spin states (i.e., eigenstates of $S_z$), we have $|x\rangle = \frac{1}{\sqrt{2}} (| - 1\rangle - | 1\rangle)$, $|y\rangle = \frac{1}{\sqrt{2}} (| - 1\rangle + | 1\rangle)$, and $|z\rangle = | 0\rangle$ [23].

The coupling between nematic tensor and OAM in $H_0$ can be more clearly seen when we carry out a gauge rotation defined by a unitary operator $U = \exp(2i\phi S_z^2)$, under which the single-particle Hamiltonian $H_0 = U H_0 U^\dagger$ in the rotating frame is explicitly expressed as

$$
\tilde{H}_0 = \frac{(i\nabla - A)^2}{2} + \frac{r^2}{2} + q S_z^2 + \sqrt{2} \Omega_R S_x,
$$

where $A = -i U^\dagger \nabla U = 2 S_z^2 \hat{e}_\phi / r$ is the synthetic gauge field on the azimuthal direction $\hat{e}_\phi$. In Eq. (2), one can clearly see the spin-nematic-OAM coupling term $\sim L_z S_z^2$ which couples the atomic quasi-OAM $L_z = -i \partial_\phi$ with one of the irreducible nematic tensors $N_{zz} = S_z^2 - 2/3$. It will play a crucial role in inducing various spin-nematic vortex states.

**Single-particle properties** — We investigate the spectrum and the eigenstates of $\tilde{H}_0$. First, we realize that $L_z$ is conserved as $[L_z, \tilde{H}_0] = 0$. Furthermore, both $L_z$ and $\tilde{H}_0$ commute with the spin parity operator

$$
P = | + 1\rangle\langle - 1 | + | - 1\rangle\langle + 1 | + | 0\rangle\langle 0 |,
$$

which satisfies $P^2 = 1$. $P$ carries a pair of eigenvalues $P = \pm 1$ distinguishing spin parity of the eigenstates. In particular, the states with even parity ($P = +$) possess the same phase on the $+1$ spin components, whereas those with odd parity ($P = -$) have a phase difference of $\pi$ on the $\pm 1$ components. It is straightforward to see that the Cartesian states $|x\rangle$ has odd spin parity, while $|y\rangle$ and $|z\rangle$ possess even spin parity. The conservation of $L_z$ ensures the quasi-OAM $l_z$ to be a good quantum number in the rotating frame, and hence the energy eigenstates can be labeled using $P$ and $l_z$ as $\tilde{\Psi}_{P = \pm, l_z} = e^{i l_z \phi} \xi_{\pm}(r) = e^{i l_z \phi} [\xi_+(r), \xi_0(r), \xi_-(r)]^T$. The spinor wave function $\xi_{\pm}(r)$ can be expanded in the Cartesian basis as

$$
\xi_+ = \sqrt{\rho(r)} \left[ i \cos \Theta(r) |y\rangle + \sin \Theta(r) |z\rangle \right],
\xi_- = \sqrt{\rho(r)} |x\rangle,
$$

where $\rho(r) = |\tilde{\Psi}(r)|^2$ is the total particle density, and $\Theta(r)$ characterizes the $r$-dependent superposition weight.

**We numerically solve the Schrödinger equation to obtain the energy spectrum and the eigenstates. Figure 2(a) displays the single-particle ground-state phase diagram in the parameter space spanned by $\Omega_0$ and $q$. Three phases I, II and III can be identified. The ground states in all three phases have even spin parity, while their quasi-OAM quantum numbers $l_z$ are 0, 1, and 2, respectively. All the phase transitions in the diagram Fig. 2(a) are of first-order since, across the phase boundary, the first order derivative of the ground-state energy with respect to $\Omega_0$ or $q$ exhibit discontinuity [20]. In each phase, we show typical energy spectrum as the inset in Fig. 2(a), where the solid dots and hollow circles distinguish the even and odd spin parity states. In Fig. 2(b), we plot the magnitude of the wave function for each bare spin component $|\xi_{0,\pm 1}|$. The first three columns represent the ground state in each phase, and the last column corresponds to an odd spin parity state $\tilde{\Psi}_{-2}$ in Phase III that lies very close to the ground state. This state will be important in our later discussion on the many-body effects in a weakly interacting condensate.

The case with vanishing quadratic Zeeman term (i.e., $q = 0$) deserves some special attention. Under this situation, the single-particle spectrum of the even-parity states is symmetric about $l_z = 1$ [20]. For $\Omega_0$ smaller than a critical value $\Omega_0 \approx 7$, the line $q = 0$ represents...
the boundary between Phase I and III [see Fig. 2(a)], on which the states $\Psi_{+/l_z}$ with $l_z = 0$ and 2 are degenerate. When $\Omega_0 > \Omega_\rho$, we enter into Phase II with $l_z = 1$. Since the term $(L_z - S_z^2) S_z^2$ in Hamiltonian (2) vanishes for $l_z = 1$ due to $S_z^2 = S_z^4$, $S_z$ is now a conserved quantity. As a result, the spinor wave function $\xi_+$ becomes an eigenstate of $S_z$ featuring $\Theta = \pi/4$.

Spin-nematic vortices — In the lab frame, different spin components of the single-particle states carry different mechanical OAM as is indicated by the numbers in the subplots of Fig. 2(b). For the spin-0 component, this is simply the quasi-OAM quantum number $l_z$; whereas for the spin-(±1) component, it is $l_z - 2$. The transformation between the wave function in the lab frame $\Psi$ and that in the rotating frame $\tilde{\Psi}$ is given by $\tilde{\Psi} = U^{\dagger}\Psi$, which explicitly leads to

$$\Psi_{+/l_z} = \sqrt{\rho(r)} e^{il_z\phi} \left(ie^{-2i\phi}\cos(\Theta)r + \sin(\Theta)r \right),$$
$$\Psi_{-/l_z} = \sqrt{\rho(r)} e^{i(l_z-2)\phi} \left|x\right>.$$  \hspace{1cm} (5)

Clearly, the ground state of Phase II, represented by $\Psi_{+,l_z=1}$, is a singular vortex as each of its bare spin components carry a finite vorticity and hence the total density vanishes at $r = 0$; while those in Phase I and III, represented by $\Psi_{+,l_z=0,2}$, are coreless vortices that contain at least one spin component with no vorticity with finite density at $r = 0$.

We investigate the spin and nematic textures by calculating the normalized spin density

$$S_\mu = \frac{\Psi^{\dagger} S_\mu \Psi}{\rho(r)},$$  \hspace{1cm} (6)

and the normalized nematic density

$$N_{\mu,\nu} = \frac{\Psi^{\dagger} N_{\mu,\nu} \Psi}{\rho(r)},$$  \hspace{1cm} (7)

where $N_{\mu,\nu} = \frac{1}{2}(S_\mu S_\nu + S_\nu S_\mu) - \frac{3}{2}\delta_{\mu\nu}$ are the symmetrized SU(3) nematic tensors with nine components by taking $\mu, \nu = x, y, z$ [9, 15, 16]. Diagonalizing the nematic density matrix $N$ results in three eigenvalues $\lambda_{1,2,3}$ characterizing the alignment axis of nematic orders. A uniaxial nematic state is characterized by $\lambda_1 \neq \lambda_2 = \lambda_3$, while for a biaxial nematic state, $N$ has two eigenvalues being equal. We remark that the Cartesian states are closely related to the SU(3) operators, which greatly facilitates the calculation of $\mathcal{S}$ and $\mathcal{N}$ [20].

Now we discuss the four low-energy states $\Psi_{+,l_z=0,1,2}$ and $\Psi_{-,l_z=2}$ represented in Fig. 2(b). The odd-parity state $\Psi_{-,l_z=2}$ is topologically trivial, since it is simply a polar state with vanishing spin density, and a fixed uniaxial direction along $N_{xz}$. For the even-parity states $\Psi_{+,l_z=0,1,2}$ which represent the ground state in the three phases, we have

$$\mathcal{S} = \{-\cos(2\phi)\sin(2\Theta), 0, 0\},$$
$$\mathcal{N} = \begin{bmatrix}
\frac{1}{3} & 0 & 0 \\
0 & -\frac{1}{2}(1 + 3\cos 2\Theta) & -\frac{1}{4}\sin(2\Theta)\sin(2\phi) \\
0 & -\frac{3}{2}\sin(2\Theta)\sin(2\phi) & -\frac{1}{4}(1 - 3\cos 2\Theta)
\end{bmatrix},$$  \hspace{1cm} (8)

where the spin vector $\mathcal{S}$ is polarized along $S_x$. The eigenvalues of the nematic matrix $\mathcal{N}$ can be obtained as $\lambda_1 = 1/3$, $\lambda_{2,3} = -1/6[1 \pm 3\sqrt{1 - \sin^2(2\Theta)\cos^2(2\phi)}]$, indicating that all three states are biaxial nematic states. Furthermore, the spin and nematic densities in Eq. (8) are not independent, and satisfy [15]

$$\frac{1}{2} \left|\mathcal{S}^2\right| + \text{Tr}[\mathcal{N}^2] = \frac{2}{3}.  \hspace{1cm} (9)$$

Substituting Eq. (8) into Eq. (9), one immediately obtains a relation

$$S_x^2 + D_{yz}^2 + (2N_{yz})^2 = 1,$$  \hspace{1cm} (10)

where $D_{yz} = N_{yy} - N_{zz} = -\cos(2\Theta)$. This relation motivates the construction of the following spin-nematic vector

$$\mathcal{Q} = \{S_x, 2N_{yz}, D_{yz}\},$$  \hspace{1cm} (11)

which forms a complete SO(3) manifold lying on a unit Bloch sphere. In fact, the vector $\mathcal{Q}$ is defined on an SU(2) group generated by $\mathcal{Q} = \{S_x, 2N_{yz}, D_{yz} = N_{yy} - N_{zz}\}$. Mathematically, the group $\mathcal{Q}$ is a type-2 subgroup of the SU(3) Lie group with the structure constant being equal to 2 [20, 24], i.e. $[2N_{yz}, S_x] = 2iD_{yz}$.

In Figure 2(c), we display the textures of the spin-nematic vector $\mathcal{Q}$ for the ground states of the three phases $\Psi_{+,l_z=0,1,2}$. One can clearly see that the transverse components of $\mathcal{Q}$ for all three states form a vortex pattern, hence the name spin-nematic vortex states.

We investigate the topological properties of the spin-nematic vortex states by examining the homotopy group [9, 18, 25, 26] of $\mathcal{Q}$. Let us first focus on the $q = 0$ case. The ground state in Phase II is a singular vortex state $\Psi_{+,l_z=1}$ satisfying $\Theta = \pi/4$ as mentioned before. Hence $\mathcal{Q}$ lies on the equator of the Bloch sphere since its $z$-component vanishes, i.e., $D_{yz} = -\cos(2\Theta) = 0$. As a result, its order manifold is reduced from SO(3) to U(1) with U(1) accounting for the azimuthal angle of the vector $\mathcal{Q}$. Since the fundamental homotopy group of U(1) is the additive integer group $\mathbb{Z}$, i.e. $\pi_1(U(1)) = \mathbb{Z}$, it allows us to depict the singular vortex by the winding number, which apparently equals to $-2$ for state $\Psi_{+,l_z=1}$. For the two coreless states $\Psi_{+,l_z=0,2}$ of Phase I and III, the vector manifold $\mathcal{Q}$ remains SO(3). However, we have the boundary condition $\Theta(r \to \infty) = \pi/4$. This condition can be clearly observed as the contribution of term $(L_z - S_z^2) S_z^2/r^2$ in Hamiltonian (2) diminishes as
Consequently, each state ($\Psi_{l_0, l_z} = 0$ or $\Psi_{l_0, l_z} = \pm 2$) covers one half of the Bloch sphere, and their topological number can thus be characterized by the skyrmion number \cite{27}

$$W = \frac{1}{4\pi} \int d^2 r \mathbf{Q} \cdot (\partial_x \mathbf{Q} \times \partial_y \mathbf{Q}), \quad (12)$$

which turns out to be $\mp 1$ for states $\Psi_{l_0, l_z} = 0, \pm 2$, respectively. Note that the coreless spin-nematic vortices defined in the type-2 subspace $\mathbf{Q}$ here are analogous to the Mermin-Ho vortex \cite{28} defined in the type-1 subspace $\mathbf{S} = \{S_x, S_y, S_z\}$. However, the two are not mathematically equivalent since the subspaces $\mathbf{S}$ and $\mathbf{Q}$ cannot be transformed into each other by SU(3) rotations \cite{24}. We note that, at finite $q$, these topological numbers are not well defined. For example, at finite $q$, the singular vortex can no longer be described by the winding number since $\pi_1(\text{SO}(3)) = 0$, and the skyrmion number of the other two coreless vortices are also in general not quantized to be integers.

**Many-body effects** — Next, we consider a weakly interacting condensate and discuss the effects of the interactions under the framework of the mean-field theory. In the lab frame, the interacting Hamiltonian of the spin-1 condensate is in the well-known form of \cite{9, 29}

$$H_{\text{int}} = \frac{1}{2} \int d^2 r \rho^2 (r) \left[ c_0 + c_2 \mathbf{S}^2 (r) \right], \quad (13)$$

where $c_0$ and $c_2$ denote the strength of the density-density and the spin-exchange interactions, respectively. We employed two different methods to obtain the mean-field ground states — the variational method and the numerical method by solving the Gross-Pitaevskii equations \cite{20}, and the results turn out to be in excellent agreement with each other. For the variational method, we assume the condensate wave function is a linear combination of the four lower-lying single-particle states shown in Fig. 2(b):

$$\tilde{\Psi} = D_0 \tilde{\Psi}_{+, 0} + D_1 \tilde{\Psi}_{+, 1} + D_+ \tilde{\Psi}_{+, 2} + D_- \tilde{\Psi}_{-, 2}, \quad (14)$$

where $D_j = 0, 1, \pm$ are variational amplitudes satisfying $\sum_j |D_j|^2 = 1$. We obtain the ground states by minimizing the total energy functional with respect to $D_j$. Here, we consider a weak ferromagnetic spin-exchange interaction by taking $c_0 = 1$ and $c_2 = -0.1c_0$, and then map out the ground-state phase diagram as is displayed in Fig. 3(a).

The main structure of the many-body phase diagram Fig. 3(a) is consistent with that of the single-particle phase diagram Fig. 2(a). In Fig. 3(a), the three phases I, II, and III are very similar to the corresponding single-particle ones in diagram Fig. 2(a) with $|D_j = 0, 1, +| = 1$ in ansatz (14), respectively, and the phase transitions among them are all of first order (solid lines). There are, however, two new many-body phases labeled as IV and V that have no counterparts in the single-particle phase diagram, and the phase transitions related to them can be either first-order (solid lines) or second-order (dashed lines), as determined by whether the first- or second-order derivatives of the ground state energy with respect to the parameters ($\Omega$ or $q$) exhibit discontinuity or not. More detailed discussions on the phase transitions and the effects of $c_2$ can be found in the Supplementary Materials \cite{20}.

These two new phases, IV and V, spontaneously break the spin parity and the rotation symmetry, respectively. Specifically, the wave function of phase IV is a superposition of states $\tilde{\Psi}_{\pm 2}$ with variational amplitudes satisfying $|D_{\pm 2}| \neq 0$ and $\theta_2 - \theta_0 = 0 \text{ or } \pi \text{ (mod } 2\pi)$. Thus, it keeps the rotational symmetry but breaks the spin-parity symmetry. Interestingly, this state exhibits vorticity in both the spin and the spin-nematic subspaces $\mathbf{S}$ and $\mathbf{Q}$ simultaneously, as are shown in Fig. 3(b1) and (b2) respectively. The breaking of the spin parity symmetry is manifested in the fact that $S_z$ is finite, i.e., unequal occupation on the bare spin-(±1) components.

The wave function of phase V is a superposition of states $\tilde{\Psi}_{+, 0}$ and $\tilde{\Psi}_{+, 2}$ with $|D_{0, +}| \neq 0$. Thus, this state maintains the spin parity symmetry, but breaks the rotational symmetry, which leads to an interesting angular striped phase \cite{30, 31}. We show the total density profile $\rho(r)$ of phase V as an inset in Fig. 3(a), where the lack of the rotational symmetry is obvious.

**Experimental observation** — Finally, let us briefly discuss the experimental detection of the spin-nematic vortex states, which can be performed either directly or indirectly. The indirect observation is to detect such features of the wave functions $\tilde{\Psi}_{l_0, l_z} = 0, 2$ (presented in Fig. 2(b)) as the core structures or the mechanical-OAM numbers. Specifically, the core structure can be obtained by the spin-selected absorption imaging; the mechanical-OAM quantum numbers can be deduced from the interference pattern after different spin components are mixed with
each other by a radio-frequency $\pi/2$ pulse [13, 14]. In contrast, the direct observation is to detect the spin-nematic textures $Q$ directly. Since the direct observation of the spin texture $S$ has been realized via the spin-sensitive dispersive imaging in a few years ago [32, 33], this technique can be easily generalized to measure $Q$ as the nematic operators $2N_y$ and $D_y$ are rotated into the measurable direction $S_z$. Practically, this rotation can be achieved by pulsing a quadratic Zeeman magnetic field which lets $Q$ evolve under the government of $\sim S_z^2$ [34], or more feasibly introducing an additional far off-resonant microwave on certain Zeeman level [35].

Summary — To summarize, we propose a scheme to couple the atomic OAM and the nematic tensor in a spin-1 cold atomic system. The ground state exhibits vorticity in a special spin-nematic subspace. Under zero quadratic field, the spin-nematic vortices can be characterized by quantized topological numbers. These features survive in the presence of weak interaction. However, the interaction may induce spontaneous symmetry breakings, and leads to a rich many-body phase diagram. Considering the spin-OAM coupling has been realized by two experimental groups very recently [13, 14, 21], we expect this work to stimulate more investigations on the spin-OAM coupled quantum gases with higher spins and nematic orders.

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In this Supplemental Materials, we provide additional information of this work. First, we provide a detailed derivation of the single-particle Hamiltonian, and then explain the symmetry and degeneracy of the single-particle spectrum at $q = 0$. Additionally, we discuss the SU(3) operators and the classification of the SU(2) subspaces, and then we show the relations between the Cartesian states and the spin and nematic densities. Furthermore, we specifically display the spin and spin-nematic orders in our single-particle and many-body phase diagrams, and show how various phase transition shown in the main text are classified. Finally, we discuss some calculation details of the numerical evolution of the coupled Gross-Pitaevskii (GP) equations.

**DERIVATION OF THE SINGLE-PARTICLE HAMILTONIAN**

We provide detailed information of our proposal in Fig. S1 as a supplement of the schematic shown in the main text, where three Laguerre-Gaussian laser beams, with optical frequency $\omega_{j=1,2,3}$ and orbit angular momentum (OAM) $h_1, h_2$ and $h_3$, respectively, propagate along the $z$-direction and shine on a quasi-2D Bose-Einstein condensate (BEC). A bias magnetic field on the $x$-direction provides a fixed quantization axis. The ground-state $^2S_{1/2}$ and the excited manifold $^2P_{1/2}$ (D1 line) and $^2P_{3/2}$ (D2 line) are coupled by the Raman beams in the way as is shown in Fig. S1(b). Note that, in principle, this scheme can be used in an arbitrary alkaline-metal atomic species. Then, the single-particle Hamiltonian reads (we set $\hbar = 1$)

$$
H_0 = h_0 + h_{\text{at}} + h_{\text{int}}
= -\frac{1}{2} \nabla^2 + \frac{1}{2} \varepsilon^2 + \sum_{j \in g} \omega_j^g |P_j^g|^2 + \sum_{j \in e} \omega_j^e |P_j^e|^2 + A f \mathbf{L} \cdot \mathbf{S} - \mathbf{d} \cdot \mathbf{E},
$$

(S1)

where $h_0$, consisting the first four terms on the second line, is the bare Hamiltonian of the single atom with $P_{j}^{g,e} = \sum_{j \in g,e} |j\rangle \langle j|$ being the projection operators of the ground-state (g) or the excited-state (e) atomic manifold, and accordingly the $\omega_j^{g,e}$ being the wave-number of the ground or excited states. The term $h_{\text{at}} = A f \mathbf{L} \cdot \mathbf{S}$ represents the fine structure spin-orbit coupling of the valence electron with $A f$ being the fine-structure interaction strength, and the last term $h_{\text{int}} = -\mathbf{d} \cdot \mathbf{E}$ represents the atom-light electric dipole interaction with $\mathbf{E}$ the electromagnetic field, and $\mathbf{d}$ the electric dipole moment of the atom.

For the Raman process with large single-photon detuning $\omega_e - \omega_g \gg \omega_L$, the excited states can be adiabatically eliminated by the second-order perturbation theory [1] and the resulting effective Hamiltonian in the ground-state manifold is in the form of

$$
h_{\text{eff}} = -P_{\text{eff}} h_{\text{int}} P_{\text{eff}}^{-1} h_{\text{int}} P_{\text{eff}} = \left[ u_{\text{s}} |\mathbf{E}|^2 + i u_{\text{e}} (\mathbf{E}^* \times \mathbf{E}) \cdot \mathbf{S} \right] P_{\text{eff}}.
$$

(S2)

$h_{\text{eff}}$ has two main effects. The first term proportional to $u_{\text{s}} \propto (|g\rangle |e\rangle)^2 / \Delta$ is the light-induced scalar shift (or ac-Stark shift) being independent on the polarization of the optical beams where $\Delta = \omega_L - (\omega_g - \omega_e)$ is the single-photon detuning; the second term with strength $u_{\text{e}} \propto A f u_{\text{s}} / \Delta$ is the light-induced vector shift. The scalar shift can be switched off as one chooses the tune-out optical frequency [2] for the Raman beams such that the scalar shifts induced by the $D_2$ and $D_1$ transitions cancel with each other.

Properly engineering the atom-light interaction, the light-induced vector shift would lead to synthetic gauge field or synthetic spin-orbit coupling [1]. Particularly for the current Raman configuration in our scheme with $\omega_{L,3}^1$ being linearly polarized along the $x$-direction, and $\omega_{L}^2$ linearly polarized along the $y$-direction, the vector shift induced by the electromagnetic field

$$
\mathbf{E} = E_1 + E_2 + E_3
= \left[ E_1 e^{i(\phi + \omega_{L}^1 t)} + E_3 e^{i(\phi + \omega_{L}^3 t)} \right] \hat{e}_x + E_2 e^{i(-\phi + \omega_{L}^2 t)} \hat{e}_y + c.c.,
$$

(S3)

...
can be easily worked out as
\[
    h_{\text{eff}} = \left(\Omega_{12} e^{-i\delta\omega_{12}t - i2i\phi} + \Omega_{23}^* e^{-i\delta\omega_{23}t - 2i\phi}\right) S_z + \text{h.c.},
\]  
where we have set \( \Omega_{ij} = -iu_iE_i^*E_j/2 \) to be the two-photon Raman frequency and \( \delta\omega_{ij} = \omega_i - \omega_j \), and the \( S_z = -(|1\rangle \langle 2| + |2\rangle \langle 3|) + \text{h.c.} \) characterizes the particle transitions in the representation of the quantized axis \( S_z \). However, in Eq. (S4), not all the transitions are allowed by the level diagram Fig. S1(b). Neglecting the forbidden transitions and the virtual photon processes (counter-rotating-wave terms), and then by simply taking \( \Omega_{12} = \Omega_{23} = \Omega_R \), we have the simplified total Hamiltonian as
\[
    H_0 = -\frac{1}{2} \nabla^2 + \frac{1}{2} r^2 + \sum_{j \in g} \omega_j^g P_j^g + \Omega_R \begin{pmatrix} 0 & e^{-2i\phi - i\delta\omega_{12}t} & 0 \\ e^{2i\phi + i\delta\omega_{12}t} & 0 & e^{2i\phi + i\delta\omega_{23}t} \\ 0 & e^{-2i\phi - i\delta\omega_{23}t} & 0 \end{pmatrix}.
\]  
Under a standard procedure, we rewrite the Hamiltonian (S5) in a rotating frame defined by the unitary operator \( U = e^{i(\delta\omega_{12}^g P_1^g + \delta\omega_{23}^g P_2^g) t} \) as
\[
    H_0 = -\frac{1}{2} \nabla^2 + \frac{1}{2} r^2 + \begin{pmatrix} q + \delta & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & q - \delta \end{pmatrix} + \Omega_R \begin{pmatrix} 0 & e^{-2i\phi} & 0 \\ e^{2i\phi} & 0 & e^{2i\phi} \\ 0 & e^{-2i\phi} & 0 \end{pmatrix},
\]  
where we have used the relations \( \delta = (\delta_{32} - \delta_{12})/2 \) and \( q = (\delta_{32} + \delta_{12})/2 \) with \( \delta_{ij} = \delta\omega_{ij}^g - (\omega_i^g - \omega_j^g) \) being the two-photon detuning. Finally, a global spin rotation \( S_x \rightarrow S_z \), \( S_z \rightarrow S_y \) and \( S_y \rightarrow S_x \) helps us to enter the commonly used \( S_z \) representation and transforms Hamiltonian (S6) into Hamiltonian (1) in the main text.

**SPECTRUM AT ZERO QUADRATIC ZEEMAN SPLITTING**

An apparent feature in the case of \( q = 0 \) is that the energy spectrum of the even-parity states are symmetric about \( l_z = 1 \). It means there is a symmetry only existing in the even-parity subspace of the Hamiltonian \( \tilde{H}_0 \) (Eq. (2) in the main text). The even-parity subspace is spanned by the even-parity Cartesian states \( |y\rangle \) and \( |z\rangle \), under which basis, \( \tilde{H}_0 \) can be written into the matrix form
\[
    \tilde{H}_0 = -\frac{\partial^2}{2\partial r^2} - \frac{\partial}{2r\partial r} + \frac{r^2}{2} - \frac{1}{2r^2} \begin{pmatrix} (l_z - 2)^2 & 0 & 0 \\ 0 & l_z^2 & 0 \\ 0 & 0 & -i \end{pmatrix} + \sqrt{2}\Omega_R \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix},
\]
where we have used the relations $\nabla^2 = \partial^2_r + \partial_r/r + \partial^2/\partial \phi^2$ and $L_z = -i\partial_\phi$, and the properties of the Cartesian states that we will discuss in details in the following section.

Considering $l_z = 1 + l'_z$, we have

$$\tilde{H}_0 = -\frac{\partial^2}{2\partial r^2} - \frac{\partial}{2r\partial r} + \frac{r^2}{2} \left[ \left( l'_z - \frac{1}{2} \right)^2 + 0 \right] + \sqrt{2}\Omega R \left[ 0 -i \right], \quad (S8)$$

and $l_z = 1 - l'_z$, which leads to

$$\tilde{H}_0 = -\frac{\partial^2}{2\partial r^2} - \frac{\partial}{2r\partial r} + \frac{r^2}{2} \left[ \left( l'_z + \frac{1}{2} \right)^2 + \frac{1}{2} \right] + \sqrt{2}\Omega R \left[ 0 -i \right]. \quad (S9)$$

The two Hamiltonians (S8) and (S9) should have the same spectrum as they are related by a unitary transformation

$$U = \begin{bmatrix} 0 & i \\ -i & 0 \end{bmatrix} \quad (S10)$$

As a result, given a state with quasi-OAM $(1 + l'_z)$, there exists a degenerate state with quasi-OAM $(1 - l'_z)$. Hence the spectrum is symmetric about $l_z = 1$.

**SU(3) OPERATORS AND SUBSPACES CLASSIFICATION**

In the main text, we defined a series of SU(3) operators including the three spin operators $S_{\mu=x,y,z}$ and nine symmetrized nematic operators $N_{\mu\nu} = \frac{1}{2}(S_\mu S_\nu + S_\nu S_\mu) - \frac{1}{2}\delta_{\mu\nu}$. Under the basis of bare spin states $|\pm 1\rangle$ and $|0\rangle$, these operators have the following explicit matrix form:

$$S_x = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \quad S_y = i \sqrt{2} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \quad S_z = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix},$$

$$N_{xx} = \frac{1}{6} \begin{pmatrix} -1 & 0 & 3 \\ 0 & 2 & 0 \\ 3 & 0 & -1 \end{pmatrix}, \quad N_{yy} = \frac{1}{6} \begin{pmatrix} -1 & 0 & -3 \\ 0 & 2 & 0 \\ -3 & 0 & -1 \end{pmatrix}, \quad N_{zz} = \frac{1}{3} \begin{pmatrix} 1 & 0 & 0 \\ 0 & -2 & 0 \\ 0 & 0 & 1 \end{pmatrix},$$

$$N_{xy} = \frac{1}{2} \begin{pmatrix} 0 & 0 & i \\ 0 & 0 & 0 \\ -i & 0 & 0 \end{pmatrix}, \quad N_{yz} = \frac{i}{2\sqrt{2}} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \quad N_{zx} = \frac{1}{2\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & -1 \\ 0 & -1 & 0 \end{pmatrix}, \quad (S11)$$

with $N_{yz} = N_{zy}$, $N_{xz} = N_{zx}$, and $N_{yz} = N_{xz}$ by definition. However, only eight of the above operators are linearly independent, and form a complete set of generators of the SU(3) Lie group laying as the mathematical foundation of the spin-1 quantum system.

The SU(3) group has a large number of SU(2) subgroups (or SU(2) subspaces) which are generated by triads of operators $\{\hat{O}_i, \hat{O}_j, \hat{O}_k\}$ satisfying cyclic commutation relation $[\hat{O}_i, \hat{O}_j] = i\epsilon_{ijk}\hat{O}_k$ where $\epsilon$ is the structure constant and $\epsilon_{ijk}$ is the Levi-Civita antisymmetric tensor. Root diagram obtained in the adjoint representation of the Cartan subalgebra provides a powerful way in identifying all the SU(2) subspaces [3]. The subspaces can and can only be classified into two types with structure constant $\alpha$ being equal to 1 and 2, respectively. The most typical type-1 subspace that has received tremendous attention is the spin subspace $S = \{S_x, S_y, S_z\}$ with $\alpha = 1$. The spin-nematic subspace $Q = \{S_x, 2N_{yz}, D_{yz} = N_{yy} - N_{zz}\}$ used in the main text is of type-2 with $\alpha = 2$. It has been proven that the SU(3) rotations can transform subspaces belonging to the same type, but cannot transform those belonging to different types. [3]

**CARTESEAN STATES AND SPIN-NEMATIC DENSITY**

As is shown in the main text, the Cartesian states $|\mu = x, y, z\rangle$ are eigenstates of the spin operators $S_\mu$ with zero eigenvalues, i.e. $S_\mu |\mu\rangle = 0$ [4]. The transformation matrix between the bare states and the Cartesian states is given by

$$U = \begin{pmatrix} -\frac{1}{\sqrt{2}} & i & 0 \\ 0 & 0 & 1 \\ \frac{1}{\sqrt{2}} & i & 0 \end{pmatrix}. \quad (S12)$$
In the Cartesian basis, the spin and nematic operators discussed above are in quite simple forms of

\[ \langle \mu | S_\eta | \nu \rangle = i \epsilon_{\mu \eta \nu}, \]  

and

\[ \langle \mu | N_{\eta \gamma} | \nu \rangle = -\frac{1}{2} (\delta_{\mu \eta} \delta_{\gamma \nu} + \delta_{\mu \gamma} \delta_{\eta \nu}) + \frac{1}{3} \delta_{\mu \nu} \delta_{\eta \gamma}, \]  

where subscript labels \{\mu, \nu, \eta, \gamma\} can take \{x, y, z\}.

Consider an arbitrary state expanded using the Cartesian basis\(|\psi\rangle = \sum_\mu m_\mu |\mu\rangle\), the expectation of the spin operators are

\[ S_\eta = \langle \psi | S_\eta | \psi \rangle = \sum_{\mu, \nu} m_\mu^* m_\nu \langle \mu | S_\eta | \nu \rangle = i \sum_{\mu, \nu} m_\mu^* m_\nu \epsilon_{\mu \eta \nu}, \]  

or more compactly:

\[ S = -i \mathbf{m}^* \times \mathbf{m}; \]  

the expectation of the spin nematic operators are

\[ N_{\eta \gamma} = \langle \psi | N_{\eta \gamma} | \psi \rangle = \sum_{\mu, \nu} m_\mu^* m_\nu \left[ -\frac{1}{2} (\delta_{\mu \eta} \delta_{\gamma \nu} + \delta_{\mu \gamma} \delta_{\eta \nu}) + \frac{1}{3} \delta_{\mu \nu} \delta_{\eta \gamma} \right] = \frac{\delta_{\eta \gamma}}{3} - \frac{1}{2} \sum_{\mu, \nu} m_\mu^* m_\nu (\delta_{\mu \eta} \delta_{\gamma \nu} + \delta_{\mu \gamma} \delta_{\eta \nu}), \]  

or more compactly

\[ N = \frac{1}{3} - \text{Re}[\mathbf{m}^* \otimes \mathbf{m}], \]  

where \( \otimes \) denotes the Kronecker product. With Eqs. (S16) and (S18), one can easily obtain the spin and nematic densities shown in the main text.

**QUANTUM PHASE TRANSITIONS**

In the main text, we show two phase diagrams (single-particle and many-body phases diagrams), and various quantum phase transitions that can be either first-order or second-order. Here, we present detailed information on the phase diagrams and the classification of the phase transitions.

Considering that we are dealing with the case at zero temperature \( T = 0 \), the ground-state energy \( E_0 \) is the quantity that we are mainly interested in. Besides, since the quantum state \( \Psi \) carries both spin and spin-nematic orders, the averaged spin \( \langle S_\mu \rangle \) and nematicity \( \langle N_{\mu \nu} \rangle \) serve as macroscopic order parameters that can be used in phase identification. The averaged spin and nematicity are defined as the spatial average of the local ones, i.e.

\[ \langle S_\mu \rangle = \int d^2r \Psi^\dagger S_\mu \Psi = \int d^2r \rho(r) S_\mu(r), \]  

and

\[ \langle N_{\mu \nu} \rangle = \int d^2r \Psi^\dagger N_{\mu \nu} \Psi = \int d^2r \rho(r) N_{\mu \nu}(r), \]
where $S_\mu$ and $N_\mu^\nu$ are the normalized spin and nematic densities defined in Eqs. (6) and (7) in the main text.

In general, a first-order phase transition is featured by a discontinuity of the first-order derivative of $E_0$, and at the same time the order parameter exhibits a sudden jump; whereas a second-order transition is continuous in the first-order derivative of $E_0$, but is discontinuous in the second-order derivative, and in the meanwhile, the order parameter goes smoothly from a finite value to zero or vice versa. Therefore, the behaviors $E_0$ and the averaged spin/spin-nematic orders help us distinguish different phases as well as the order of the phase transition.

For the single-particle phase diagram shown in Fig. 2(a) in the main text, the phases I, II and III feature vanishing total spins $|\langle S \rangle|$ but non-vanishing averaged nematic order. In Figs. S2(a) and (b), we reprint the single-particle phase diagram with background colors showing the variations of the longitudinal nematic order $\langle D_{yz} \rangle$ and the ground-state energy $E_0$, respectively. One can observe that the nematic order $\langle D_{yz} \rangle$ exhibits sudden jumps across the phase boundaries indicating all the transitions among phases I, II and III are of first order (labeled by solid lines). The order of the phase transitions are confirmed by examining the behavior of $E_0$ and $\partial_q E_0$ as functions of $q$, as shown in Fig. S2(c). In Fig. S2(c), the lines with solid markers are plotted in the case of $\Omega_0 = 2$ where the transition I-III occurs at $q = 0$, and the lines with hollow markers are plotted in the case of $\Omega_0 = 10$ where two transitions III-II and II-I occur at $q \approx \pm 1$, respectively. The two cases are visually indicated by the two vertical dot-dashed lines in Fig. S2(b). It is clearly shown in the Fig. S2(c) that the energy $E_0$ varies continuously, but its first-order derivative $\partial_q E_0$ shows discontinuous jumps at the phase boundaries.

Similar analyses are performed on the many-body phase diagram displayed in Fig. S3, where subfigures (a)-(c) show the dependence of $\langle D_{yz} \rangle$, the total spin $|\langle S \rangle| = \sqrt{(S_x)^2 + (S_y)^2 + (S_z)^2}$, and $E_0$ on the phase plane $\Omega_0$-$q$. The two emergent new phases IV and V are ferromagnetic with non-vanishing total spin, i.e. $|\langle S \rangle| \neq 0$. As mentioned in the main text, these two new phases break different symmetries (Phase IV keeps the rotational symmetry but breaks the spin-parity symmetry; whereas Phase V keeps the spin-parity symmetry but breaks the rotational symmetry), and hence the phase transition between them is of first-order (denoted by solid lines), as confirmed by a sudden jump of the first-order derivative $\partial_q E_0$ across the phase boundary (not shown in the Fig. S3). In contrast, the phase transitions between the symmetry preserved phases (I, II and III) and the symmetry broken phases (IV and V) are all of the second order (denoted by dashed lines). We examine these transitions by tracking the variational amplitudes $|D_{0,1,+,,-}|$ (upper panel of Fig. S3(c)), the averaged spin and spin-nematic orders (lower panel of Fig. S3(c)) and the energy behaviors (Fig. S3(f)) at $\Omega_0 = 2.5$ (indicated by the vertical dot-dashed line in Fig. S3(c)), where three transitions IV-III, III-V, and V-I occur as $q$ is ascendingly swept. The second-order transitions are clearly demonstrated in Fig. S3(f) as the second-order derivative $\partial^2_q E_0$ is the lowest order of derivatives that exhibits a discontinuity at the phase boundary, and at the same time the total spin order $|\langle S \rangle|$ varies smoothly from a finite value to zero or from zero to a finite value.

Additionally, we discuss the phase dependence on the spin-exchange interaction strength $c_2$, which is not shown in the main text. To this end, we fix $\Omega_0 = 2.5$, and sweep $c_2/c_0$ and $q$ around zero to some extent. The resulting phase diagram is plotted in Fig. S3(d). The major feature that one can immediately observe in the phase diagram Fig. S3(d) is that the emergent phases IV and V only exist in the region $c_2 < 0$, and their phase areas diminish as $c_2$ increases from the negative to zero. This phenomenon can be understood by the fact that, for a conventional spin-1 BEC, the ferromagnetic interaction $c_2 < 0$ always favors ferromagnetic states, whereas the anti-ferromagnetic interaction $c_2 > 0$
FIG. S3: Many-body phase diagrams and phase transitions. (a)-(c) Many-body phase diagrams on the $\Omega_0$-$q$ plane with fixed $c_2/c_0 = -0.1$, where the background colors in (a), (b) and (c) denote the averaged longitudinal nematic order $\langle D_{yz} \rangle$, total spin order $\langle S \rangle = [(S_x)^2 + (S_y)^2 + (S_z)^2]^{1/2}$ and the ground-state energy $E_0$, respectively. (d) Many-body phase diagram and dependence of the $\langle D_{yz} \rangle$ on the $q$-$c_2/c_0$ plane, where $\Omega_0 = 2.5$ is fixed. In all the phase diagrams (a)-(d), the black solid and the black dashed lines indicate the first- and the second-order phase boundaries, respectively. (e) Variations of the variational amplitudes $|D|$, and the total spin and nematic orders on $q$. Upper panel: dependence of the variational amplitudes $|D|_{0,1,2,3,\ldots}$. Lower panel: dependence of the total spin and spin-nematicity $\langle S \rangle$, $\langle |S| \rangle$, and $\langle |D_{yz}| \rangle$. (f) Ground-state energy $E_0$ and its first $\partial_q E_0$ and second derivatives $\partial_q^2 E_0$, where $\partial_q E_0$ and $\partial_q^2 E_0$ exhibit discontinuity at the first- and the second-order phase boundaries, respectively. Insets: a closed look at the $\partial_q E_0$ in the regime $q \in [-0.45, -0.25]$. In subfigures (e) and (f), we take $\Omega_0 = 2.5$ and $c_2 = -0.1c_0$ as is indicated by the green dot-dashed line in subfigure (c).

prefers the anti-ferromagnetic or polar states. Therefore, our single-particle states I and III with vanishing total spin are more favored by the anti-ferromagnetic interaction $c_2 > 0$, and the emergent phases IV and V with non-vanishing total spin are more preferred by the ferromagnetic one with $c_2 < 0$.

GROSS-PITEAVSKII EQUATIONS

As mentioned in the main text, we use two different methods to obtain the many-body ground-state phase diagrams — variational method and numerically solving the Gross-Pitaevskii (GP) equations, and the results obtained by the two methods are in good agreement with each other. Particularly for the latter method, we first derive the GP equations from the total Hamiltonian $H = H_0 + H_{int}$, where $H_0$ and $H_{int}$ are single-particle and interacting Hamiltonian shown as Eq. (1) and Eq. (13) in the main text. In the lab frame, the GP equations are explicitly written in the bare basis as

\[ i\partial_t \psi_1 = \left( -\frac{\nabla^2}{2} + \frac{r^2}{2} + q + \delta + c_0 \rho \right) \psi_1 + \left( \Omega_R e^{-2i\phi} + \frac{c_2}{\sqrt{2}} \rho S_z \right) \psi_0, \]

\[ i\partial_t \psi_0 = \left( -\frac{\nabla^2}{2} + \frac{r^2}{2} + c_0 \rho \right) \psi_0 + \left( \Omega_R e^{2i\phi} + \frac{c_2}{\sqrt{2}} \rho S_+ \right) \psi_1 + \left( \Omega_R e^{2i\phi} + \frac{c_2}{\sqrt{2}} \rho S_- \right) \psi_{-1}, \]

\[ i\partial_t \psi_{-1} = \left( -\frac{\nabla^2}{2} + \frac{r^2}{2} + q - \delta + c_0 \rho \right) \psi_{-1} + \left( \Omega_R e^{-2i\phi} - \frac{c_2}{\sqrt{2}} \rho S_z \right) \psi, \]

where $\rho(r) = |\Psi(r)|^2$ is the total density, $S$ is the normalized spin density defined in Eq.(6) in the main text, with $S_\pm = S_x \pm iS_y$. Then, the many-body ground states $\Psi$ can be obtained by propagating the GP equations in imaginary time, which is commonly called the imaginary-time evolution. As for the time and space discretization, we deal with
the temporal propagation using the time-splitting method [5], and employ the pseudo-spectral method and the finite difference method to deal with the kinetic term $-\nabla^2/2$ and the other non-kinetic terms, respectively.

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