Ground states of spin-2 condensates in an external magnetic field

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The possible ground states of spin-2 Bose-Einstein condensates in an external magnetic field are obtained analytically and classified systematically according to the population of the condensed atoms at the hyperfine sublevels. It is shown that the atoms can populate simultaneously at four hyperfine sublevels in a weak magnetic field with only the linear Zeeman energy, in contrast to that in a stronger magnetic field with the quadratic Zeeman energy, where condensed atoms can at most populate at three hyperfine sublevels in the ground states. Any spin configuration we obtained will give a closed subspace in the order parameter space of the condensates.

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I. INTRODUCTION

Recently, spinor Bose-Einstein condensates (BECs) have received much attention in both experimental [1–4] and theoretical studies [5–8]. Spinor BECs have internal degrees of freedom because of the hyperfine spin of the atoms, which liberate a rich variety of phenomena such as spin domains and textures.

The ground states of spinor condensates in an external magnetic field are an interesting problem because of the constraint of both the atom number and total spin conservation. For the spin-1 condensates, the mean-field ground states were investigated in the pioneering work of Refs. [2,5,6] and became a hot topic later [9–11]. For the spin-2 condensates in a weak magnetic field with only the linear Zeeman energy, the mean-field ground states were first studied in Refs. [12,13]. The phase diagram was divided into three parts: the ferromagnetic, the polar, and the cyclic phases. The cyclic phase is absent for the spin-1 condensates and has drawn increasing attention. Many phenomena were found in this phase such as stable fractional vortices [14], vortex lattice transition [15], long-range order, kinks and roughening transition [16], and so on.

In Refs. [12,13], only some possible ground states of the spin-2 condensates in a weak magnetic field were given for the cyclic phase. But until now, the systematical classification of the cyclic ground-state configurations have been absent to our knowledge, and in this article, we will obtain analytically and classified systematically the possible ground-state spin structure according to the population of the condensed atoms at the hyperfine sublevels. Very recently, the ground states of spin-2 condensates in a stronger magnetic field with the quadratic Zeeman energy were obtained and classified according to the determinant of the coefficient matrix [17,18]. In this article, we will obtain these states in a simple way according to the atom population and classified systematically in terms of the singlet pair amplitude. It is shown that the condensed atoms can populate simultaneously at four hyperfine sublevels in the weak magnetic field where the linear Zeeman energy dominates, while atoms can at most populate at three hyperfine sublevels in a stronger magnetic field with the quadratic Zeeman energy.

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The article is organized as follows: In Sec. II, we introduce the model of spin-2 condensates in an external magnetic field. In Sec. III, the possible ground states in a weak magnetic field with only the linear Zeeman energy are shown. The effects of the quadratic Zeeman energy are analyzed in Sec. IV. Finally, we give conclusions and some remarks on our results in Sec. V. In the appendix, the possible ground states of the cyclic phase with only the linear Zeeman energy term are derived analytically in detail according to the population of the atoms.

II. THE MODEL

The effective low-energy Hamiltonian of a spin-f Bose gas was derived by Ho [5]. For spin-2, it reads [12]

$$\hat{H} = \int d\mathbf{r}\left\{\hat{\psi}_a^\dagger\left[-\frac{\hbar^2\nabla^2}{2M}+V_{trap}\right]\hat{\psi}_a+\frac{1}{2}c_0\hat{\psi}_a^\dagger\hat{\psi}_b^\dagger\hat{\psi}_b\hat{\psi}_a\right\} + \frac{1}{2}\int d\mathbf{r}\{c_1[\hat{\psi}_{a'}^\dagger(\vec{\mathbf{F}})_{a'b'}\hat{\psi}_{b'}]\cdot[\hat{\psi}_a^\dagger(\vec{\mathbf{F}})_{ab}\hat{\psi}_b] + c_2[\hat{\psi}_{a'}(\mathbf{A})_{a'b'}\hat{\psi}_{b'}]^\dagger[\hat{\psi}_a(\mathbf{A})_{ab}\hat{\psi}_b]\}, \quad (1)$$

where $\hat{\psi}_a$ $(a=-2,\ldots,2)$ is the hyperfine atomic field operator, V_{trap} is the trap potential, and repeated indices summate; $c_0=(4g_2+3g_4)/7, c_1=(g_4-g_2)/7$, and $c_2=(7g_0-10g_2+3g_4)/35$, where $g_F=4\pi\hbar^2a_F/M$ $(F=0,2,4)$, with a_F being the s-scattering lengths in the total spin F channel and M being the atomic mass. $\vec{\mathbf{F}}$ is the spin-2 matrix [13]. $\mathbf{A}$ is associated with the singlet pair amplitude [12], and the matrix element $(\mathbf{A})_{ab}=(-1)^a\delta_{a+b,0}$.

The external magnetic field $\mathbf{B}$ is along the z direction, which is taken to be the quantization axis. It introduces the Zeeman splitting energy term [9,11,12,17]

$$\hat{H}_{ZM}(B)=\int d\mathbf{r}\{-p_0\hat{\psi}_a^\dagger(\mathbf{F}_z)_{ab}\hat{\psi}_b+q_0\hat{\psi}_a^\dagger(\mathbf{F}_z)_{ab}^2\hat{\psi}_b\}, \quad (2)$$

where p_0 and q_0 measure the linear and quadratic Zeeman effects, respectively.

If the ground state is unfragmented, the field operator becomes a c number $\Psi_a=\langle\hat{\psi}_a\rangle=\sqrt{n}\zeta_a$, where n is the density and ζ is a normalized spinor, $\zeta^+\zeta=\zeta_a^*\zeta_a=1$. For the homogeneous condensates, the spin configuration ζ is determined by the energy functional

$$\varepsilon(\zeta)=\frac{c_0n^2}{2}+\frac{c_1n^2}{2}\langle\vec{\mathbf{F}}\rangle^2+\frac{c_2n^2}{2}|\Theta|^2-p_0n\langle\mathbf{F}_z\rangle+q_0n\langle\mathbf{F}_z^2\rangle, \tag{3}$$

where $\Theta=\zeta^T(\mathbf{A})\zeta=\zeta_a(\mathbf{A})_{ab}\zeta_b=2\zeta_2\zeta_{-2}-2\zeta_1\zeta_{-1}+\zeta_0^2$ is related to the singlet pair amplitude. In the ground state, the magnetization of the system must be aligned with the external field [12], implying that $\langle\mathbf{F}_+\rangle=0$, where $\mathbf{F}_+=\mathbf{F}_x+i\mathbf{F}_y$. Then Eq. (3) becomes

$$\varepsilon(\zeta)=\frac{c_0n^2}{2}+\frac{c_1n^2}{2}\langle\mathbf{F}_z\rangle^2+\frac{c_2n^2}{2}|\Theta|^2-p_0n\langle\mathbf{F}_z\rangle+q_0n\langle\mathbf{F}_z^2\rangle. \tag{4}$$

Minimizing Eq. (4) under the constraint $\zeta^+\zeta=1$, we obtain the stationary Gross-Pitaevskii equations of the spinor as follows:

$$(4q+2\gamma-\lambda)\zeta_2+c_2\Theta\zeta_{-2}^*=0, \tag{5}$$
$$(q+\gamma-\lambda)\zeta_1-c_2\Theta\zeta_{-1}^*=0, \tag{6}$$
$$c_2\Theta\zeta_0^*-\lambda\zeta_0=0, \tag{7}$$
$$(q-\gamma-\lambda)\zeta_{-1}-c_2\Theta\zeta_1^*=0, \tag{8}$$
$$(4q-2\gamma-\lambda)\zeta_{-2}+c_2\Theta\zeta_2^*=0, \tag{9}$$

where $p=p_0/n, q=q_0/n$ and $\gamma=c_1\langle\mathbf{F}_z\rangle-p;\lambda=\mu-c_0$, and the chemical potential μ is a Lagrangian multiplier for the normalization of the spinor.

The ground states can be obtained by solving the preceding five equations under the constraints of no transverse magnetization, namely,

$$\langle\mathbf{F}_+\rangle=2\zeta_2^*\zeta_1+\sqrt{6}\zeta_1^*\zeta_0+\sqrt{6}\zeta_0^*\zeta_{-1}+2\zeta_{-1}^*\zeta_{-2}=0, \tag{10}$$

the normalization condition,

$$|\zeta_2|^2+|\zeta_1|^2+|\zeta_0|^2+|\zeta_{-1}|^2+|\zeta_{-2}|^2=1, \tag{11}$$

and the total spin conservation,

$$\langle\mathbf{F}_z\rangle=2|\zeta_2|^2-2|\zeta_{-2}|^2+|\zeta_1|^2-|\zeta_{-1}|^2\equiv m. \tag{12}$$

In the weak external magnetic field, the Zeeman shifts are substantially smaller than the hyperfine splitting, and the quadratic shifts are typically smaller than the linear shifts [11]. Thus we proceed first with only the linear Zeeman term, and then the effects of the quadratic shifts are analyzed.

III. GROUND STATES IN THE WEAK MAGNETIC FIELD

In the absence of the quadratic Zeeman term, that is, $q=0$, the stationary Eqs. (5)–(9) can be written in a matrix form:

$$[c_1\langle\mathbf{F}_z\rangle-p](\mathbf{F}_z)\zeta-\lambda\zeta+c_2\Theta(\mathbf{A})\zeta^*=0. \tag{13}$$

Multiplying Eq. (13) with ζ^+ from the left-hand side, we have

$$c_1\langle\mathbf{F}_z\rangle^2+c_2|\Theta|^2-p\langle\mathbf{F}_z\rangle-\lambda=0, \tag{14}$$

where $\Theta^*=\zeta^+(\mathbf{A})\zeta^*$. Multiplying Eq. (13) with $\zeta^T(\mathbf{A})$ from the left-hand side, we get

$$\Theta(\lambda-c_2)=0, \tag{15}$$

where we have used the relation $\zeta^T(\mathbf{A})(\mathbf{F}_z)\zeta=0$ and $(\mathbf{A})(\mathbf{A})=1$.

We follow the terminology of Ref. [12]. For the polar phase, $\Theta\neq0$, that is, $\lambda=c_2$, namely, the chemical potential $\mu=c_0+c_2$. Then one has [12]

$$(\mathbf{F}_z^2)\zeta=\frac{c_2^2(1-|\Theta|^2)}{[c_1\langle\mathbf{F}_z\rangle-p]^2}\zeta, \tag{16}$$

showing that ζ is an eigenstate of $(\mathbf{F}_z^2)$ with possible eigenvalues 0, 1, 4, denoted as P0, P1, and P, respectively:

$$\text{P0: }\zeta^T=e^{i\alpha_0}(0,0,1,0,0), \tag{17}$$

with the total spin $m=0$,

$$\text{P1: }\zeta^T=\left(0,e^{i\alpha_1}\sqrt{\frac{1}{2}+\frac{m}{2}},0,e^{i\alpha_{-1}}\sqrt{\frac{1}{2}-\frac{m}{2}},0\right), \tag{18}$$

with $m=p/(c_1-c_2)$, and

$$\text{P: }\zeta^T=\left(e^{i\alpha_2}\sqrt{\frac{1}{2}+\frac{m}{4}},0,0,0,e^{i\alpha_{-2}}\sqrt{\frac{1}{2}-\frac{m}{4}}\right), \tag{19}$$

with $m=4p/(4c_1-c_2)$; α_i are arbitrary phases. All these states satisfy $\langle\mathbf{F}_+\rangle=0$ and $\Theta\neq0$.

If $\Theta=0$, Eq. (13) minus Eq. (14), multiplying with ζ from the right-hand side, yields

$$[c_1\langle\mathbf{F}_z\rangle-p][(\mathbf{F}_z)-\langle\mathbf{F}_z\rangle]\zeta=0. \tag{20}$$

For the ferromagnetic phase, $(\mathbf{F}_z)\zeta=\langle\mathbf{F}_z\rangle\zeta$, ζ is the eigenstate of $\mathbf{F}_z$, which leads to the states F2, F1, F-1, F-2, respectively:

$$\text{F2: }\zeta^T=e^{i\alpha_2}(1,0,0,0,0),\ \ m=2, \tag{21}$$
$$\text{F1: }\zeta^T=e^{i\alpha_1}(0,1,0,0,0),\ \ m=1, \tag{22}$$
$$\text{F-1: }\zeta^T=e^{i\alpha_{-1}}(0,0,0,1,0),\ \ m=-1, \tag{23}$$
$$\text{F-2: }\zeta^T=e^{i\alpha_{-2}}(0,0,0,0,1),\ \ m=-2. \tag{24}$$

For the cyclic phase, it is required that $\langle\mathbf{F}_z\rangle=p/c_1$, compared with the ferromagnetic phase. When $m=\pm2,\pm1$, some of the cyclic states can reduce to the ferromagnetic states. From Eq. (13), we have $\mu=c_0$.

In the appendix, we obtain analytically and classify systematically the possible cyclic ground-state configurations according to the population of the condensed atoms at the hyperfine sublevels. It is shown that the condensed atoms can populate simultaneously at four hyperfine sublevels in the weak magnetic field with only the linear Zeeman energy.

IV. GROUND STATES WITH THE QUADRATIC ZEEMAN ENERGY

In the presence of the quadratic Zeeman term, we have to solve the five stationary Gross-Pitaevskii equations [Eqs. (5)–(9)], under the constraints of no transverse magnetization, namely, Eq. (10), the normalization condition [Eq. (11)], and the total spin [Eq. (12)]. We follow the classification principle in the case of no quadratic Zeeman energy. For the polar states, $\Theta=2\zeta_2\zeta_{-2}-2\zeta_1\zeta_{-1}+\zeta_0^2\neq0$. To satisfy this condition and Eq. (10), there are four possibilities: (1) $\zeta_0\neq0$; (2) $\zeta_1\neq0$ and $\zeta_{-1}\neq0$; (3) $\zeta_2\neq0$ and $\zeta_{-2}\neq0$; or (4) $\zeta_2\neq0$, $\zeta_{-2}\neq0$, and $\zeta_0\neq0$:

1. Obviously, we have the state P0, $\zeta^T = e^{i\alpha_0}(0,0,1,0,0)$, with the chemical potential $\mu = c_0 + c_2$.

2. We have $\Theta = -2\zeta_1\zeta_{-1}$ and two stationary equations

$$(q+\gamma-\lambda)\zeta_1 + 2c_2|\zeta_{-1}|^2\zeta_1 = 0, \tag{25}$$

$$(q-\gamma-\lambda)\zeta_{-1} + 2c_2|\zeta_1|^2\zeta_{-1} = 0. \tag{26}$$

The two equations and the normalization condition [Eq. (11)] yield $\lambda = q + c_2$. Then the preceding two equations give

$$\zeta^T = \left(0, e^{i\alpha_1}\sqrt{\frac{1}{2}+\frac{\gamma}{2c_2}}, 0, e^{i\alpha_{-1}}\sqrt{\frac{1}{2}-\frac{\gamma}{2c_2}}, 0\right). \tag{27}$$

The total spin $\langle \mathbf{F}_z\rangle = |\zeta_1|^2 - |\zeta_{-1}|^2 = \frac{\gamma}{c_2} = c_1/c_2\langle \mathbf{F}_z\rangle - p/c_2$, namely, $m = p/(c_1 - c_2)$. Thus this state is the same with P1, but with the different chemical potential $\mu = c_0 + c_2 + q$.

3. In the same way, we obtain the same state P, with the different chemical potential $\mu = c_0 + c_2 + 4q$.

4. In this case, we can only follow the result of Eqs. (103)–(106) of Ref. [17].

For the ferromagnetic and cyclic states, $\Theta = 0$. If all the atoms populate at only one hyperfine sublevel, we have the same four states F2, F1, F-1, and F-2. If the atoms populate at two hyperfine sublevels, we have two possible configurations: (1) $\zeta_2 \neq 0$ and $\zeta_{-1} \neq 0$ or (2) $\zeta_{-2} \neq 0$ and $\zeta_1 \neq 0$:

1. We have two stationary equations

$$(4q + 2\gamma - \lambda)\zeta_2 = 0, \tag{28}$$

$$(q - \gamma - \lambda)\zeta_{-1} = 0, \tag{29}$$

namely,

$$\lambda = 4q + 2\gamma, \tag{30}$$

$$\lambda = q - \gamma; \tag{31}$$

then we have $\lambda = 2q$ and $\gamma = -q = c_1\langle \mathbf{F}_z\rangle - p$, which yield $m = p - q/c_1$. From the normalization [Eq. (11)] and the total spin [Eq. (12)], we obtain the same state with only the linear Zeeman term

$$\zeta^T = \frac{1}{\sqrt{3}}[\sqrt{1+m}e^{i\theta_2}, 0, 0, \sqrt{2-m}e^{i\theta_{-1}}, 0] \tag{32}$$

but with different total spin m and the chemical potential $\mu = c_0 + 2q$.

2. In the same way, we obtain the state

$$\zeta^T = \frac{1}{\sqrt{3}}[0, \sqrt{2+m}e^{i\theta_1}, 0, 0, \sqrt{1-m}e^{i\theta_{-2}}], \tag{33}$$

with $m = (p+q)/c_1$ and $\mu = c_0 + 2q$.

If atoms populate at only three hyperfine sublevels, basing on the analysis in the appendix, we have $\zeta_2 \neq 0$, $\zeta_{-2} \neq 0$, and $\zeta_0 \neq 0$. Then the stationary equations reduce to

$$\lambda = 0, \tag{34}$$

$$4q + 2\gamma = 0, \tag{35}$$

$$4q - 2\gamma = 0, \tag{36}$$

namely, $\mu = c_0$ and $q = 0 = \gamma = c_1\langle \mathbf{F}_z\rangle - p$, which yield $m = p/c_1$. Thus the state reduces to that with only the linear Zeeman energy, namely, in the presence of the quadratic Zeeman energy, the atoms cannot populate at the three hyperfine sublevels in the cyclic phase. If atoms populate at four or five hyperfine sublevels, we also have $q = 0$ and obtain the same states in the absence of quadratic Zeeman energy.

Finally, we want to point out that for alkali-metal condensates, the linear and quadratic Zeeman energy may also be engineered by using off-resonant microwave fields that generate electromagnetically induced level splittings [19], allowing essentially arbitrary experimentally prepared level shifts for p and q [11]. If the quadratic Zeeman energy absolutely dominates, that is, $p = 0$, only polar states P0, as in Eq. (106) of Ref. [17], and the cyclic states [Eqs. (32) and (33)] exist.

V. CONCLUSION AND REMARKS

In this article, we obtain analytically and classify systematically the possible ground states of spin-2 BECs in an external magnetic field according to the population of the condensed atoms at hyperfine sublevels. It is shown that the atoms can populate simultaneously at four hyperfine sublevels in a weak magnetic field with only the linear Zeeman energy, in contrast to a stronger magnetic field with the quadratic Zeeman energy, where condensed atoms can at most populate at three hyperfine sublevels in the ground states. The properties of the spinor condensates in a weak or stronger magnetic field should be different. In the weak magnetic field, the linear Zeeman energy is dominant, but in the stronger magnetic field, the quadratic Zeeman energy dominates.

The rotational symmetry group SO(3) of the order parameter of the spinor condensates is reduced to SO(2) in the presence of the external magnetic field. Any two ground states we obtained cannot transform by spin rotation. Thus any spin configuration we obtained will give a closed subspace in the order parameter space of the condensates.

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APPENDIX

For all the possible states in the cyclic phase, the following four equations must be satisfied:

$$\langle \mathbf{F}_+\rangle = 2\zeta_2^*\zeta_1 + \sqrt{6}\zeta_1^*\zeta_0 + \sqrt{6}\zeta_0^*\zeta_{-1} + 2\zeta_{-1}^*\zeta_{-2} = 0, \tag{A1}$$

$$\Theta = 2\zeta_2\zeta_{-2} - 2\zeta_1\zeta_{-1} + \zeta_0^2 = 0, \tag{A2}$$

the normalization condition,

$$|\zeta_2|^2 + |\zeta_1|^2 + |\zeta_0|^2 + |\zeta_{-1}|^2 + |\zeta_{-2}|^2 = 1, \tag{A3}$$

and the total spin,

$$\langle \mathbf{F}_z\rangle = \frac{p}{c_1} = 2|\zeta_2|^2 - 2|\zeta_{-2}|^2 + |\zeta_1|^2 - |\zeta_{-1}|^2 = m, \tag{A4}$$

where the modulus $|\zeta_i| \geqslant 0$ is a real number. This is a simple but very important criterion in the following analysis.

A. All atoms populate at only one hyperfine sublevel

We then obtain four possible states, F2, F1, F-1, and F-2.

B. Atoms populate at only two hyperfine sublevels

To satisfy Eqs. (A1) and (A2), there are only two possible configurations: (1) $\zeta_2 \neq 0$ and $\zeta_{-1} \neq 0$ or (2) $\zeta_{-2} \neq 0$ and $\zeta_1 \neq 0$.

1. We have

$$2|\zeta_2|^2 - |\zeta_{-1}|^2 = m, \tag{A5}$$

$$|\zeta_2|^2 + |\zeta_{-1}|^2 = 1; \tag{A6}$$

then we obtain

$$\zeta^T = \frac{1}{\sqrt{3}}[\sqrt{1+m}e^{i\theta_2},0,0,\sqrt{2-m}e^{i\theta_{-1}},0]. \tag{A7}$$

This state exists only for $-1 \leqslant m \leqslant 2$. When $m = 2$, we have the state F2, and when $m = -1$, we have the state F-1.

2. In the same way, we obtain

$$\zeta^T = \frac{1}{\sqrt{3}}[0,\sqrt{2+m}e^{i\theta_1},0,0,\sqrt{1-m}e^{i\theta_{-2}}] \tag{A8}$$

for $-2 \leqslant m \leqslant 1$.

C. Atoms populate at only three hyperfine sublevels

To satisfy Eq. (A2), it seems that there are two possible configurations: (1) $\zeta_2 = 0$ and $\zeta_{-2} = 0$ or (2) $\zeta_1 = 0$ and $\zeta_{-1} = 0$.

1. Equation (A1) reduces to

$$|\zeta_{-1}| = -|\zeta_1|e^{i(2\theta_0-\theta_1-\theta_{-1})}, \tag{A9}$$

namely, $2\theta_0 - \theta_1 - \theta_{-1} = \pi$, but Eq. (A2) reduces to

$$2|\zeta_1||\zeta_{-1}| = |\zeta_0|^2 e^{i(2\theta_0-\theta_1-\theta_{-1})}, \tag{A10}$$

namely, $2\theta_0 - \theta_1 - \theta_{-1} = 0$, which shows the impossibility of this state.

2. Equation (A1) has been satisfied, and Eq. (A2) reduces to

$$|\zeta_0|^2 = -2|\zeta_2||\zeta_{-2}|e^{i(\theta_2+\theta_{-2}-2\theta_0)}, \tag{A11}$$

namely, $\theta_2 + \theta_{-2} - 2\theta_0 = \pi$, and $|\zeta_0|^2 = 2|\zeta_2||\zeta_{-2}|$. We also have $\theta_{-2} - \theta_0 = \pi - (\theta_2 - \theta_0)$.

Equation (A3) reduces to

$$(|\zeta_2| + |\zeta_{-2}|)^2 = 1, \tag{A12}$$

namely, $|\zeta_2| + |\zeta_{-2}| = 1$, and Eq. (A4) reduces to

$$|\zeta_2|^2 - |\zeta_{-2}|^2 = |\zeta_2| - |\zeta_{-2}| = \frac{m}{2}. \tag{A13}$$

We then obtain $|\zeta_2| = 1/2(1 + m/2)$, $|\zeta_{-2}| = 1/2(1 - m/2)$, $|\zeta_0| = 1/2\sqrt{2 - m^2/2}$, and

$$\zeta^T = \frac{1}{2}e^{i\theta_0}\left[\left(1+\frac{m}{2}\right)e^{i(\theta_2-\theta_0)}, 0, \sqrt{2-\frac{m^2}{2}}, 0, -\left(1-\frac{m}{2}\right)e^{-i(\theta_2-\theta_0)}\right], \tag{A14}$$

which is just as presented in Ref. [12].

D. Atoms populate at only four hyperfine sublevels

1. For $\zeta_0 = 0$, Eq. (A1) reduces to

$$|\zeta_2||\zeta_1| = -|\zeta_{-1}||\zeta_{-2}|e^{i(\theta_2+\theta_{-2}-\theta_1-\theta_{-1})}, \tag{A15}$$

namely, $\theta_2 + \theta_{-2} - \theta_1 - \theta_{-1} = \pi$, and Eq. (A2) reduces to

$$|\zeta_2||\zeta_{-2}| = |\zeta_1||\zeta_{-1}|e^{-i(\theta_2+\theta_{-2}-\theta_1-\theta_{-1})}, \tag{A16}$$

namely, $\theta_2 + \theta_{-2} - \theta_1 - \theta_{-1} = 0$. These two equations cannot be satisfied simultaneously, except that (1) $|\zeta_1| = 0$ and $|\zeta_{-2}| = 0$ and (2) $|\zeta_{-1}| = 0$ and $|\zeta_2| = 0$, which are just the two states in Sec. II.

2. For $\zeta_2 = 0$, Eq. (A2) reduces to

$$|\zeta_0|^2 = 2|\zeta_1||\zeta_{-1}|e^{i(\theta_1+\theta_{-1}-2\theta_0)}, \tag{A17}$$

namely, $\theta_1 + \theta_{-1} - 2\theta_0 = 0$, and

$$|\zeta_0|^2 = 2|\zeta_1||\zeta_{-1}|. \tag{A18}$$

We also have $\theta_1 - \theta_0 = -(\theta_{-1} - \theta_0)$, which reduces Eq. (A1) to

$$\sqrt{6}|\zeta_0|(|\zeta_1| + |\zeta_{-1}|) = -2|\zeta_{-1}||\zeta_{-2}|e^{i(\theta_{-2}+\theta_0-2\theta_{-1})}, \tag{A19}$$

namely, $\theta_{-2} + \theta_0 - 2\theta_{-1} = \pi$, and

$$\sqrt{6}|\zeta_0|(|\zeta_1| + |\zeta_{-1}|) = 2|\zeta_{-1}||\zeta_{-2}|. \tag{A20}$$

We also have $\theta_{-2} - \theta_0 = \pi + 2(\theta_{-1} - \theta_0)$.

Substituting Eq. (A18) into Eq. (A3) yields

$$(|\zeta_1| + |\zeta_{-1}|)^2 + |\zeta_{-2}|^2 = 1. \tag{A21}$$

Thus we take

$$|\zeta_{-2}| = \cos\alpha,\ |\zeta_1| = \sin\alpha\sin^2\beta,\ |\zeta_{-1}| = \sin\alpha\cos^2\beta, \tag{A22}$$

where $\sin\alpha, \cos\alpha, \sin\beta, \cos\beta$ are all nonzero. We then obtain

$$|\zeta_0| = \sqrt{2}\sin\alpha\sin\beta\cos\beta. \tag{A23}$$

Substituting Eqs. (A22) and (A23) into Eq. (A20), we have

$$\sqrt{3}\sin\alpha\sin\beta = \cos\alpha\cos\beta. \tag{A24}$$

Substituting Eqs. (A22) and (A23) into Eq. (A4), we have

$$-2\cos^2\alpha + \sin^2\alpha(\sin^2\beta - \cos^2\beta) = m, \tag{A25}$$

and

$$\sin^2\alpha = (m+2)/(2\sin^2\beta + 1).$$

Substituting it into Eq. (A24), we have

$$2\sin^4\beta + (2m+3)\sin^2\beta + m + 1 = 0, \tag{A26}$$

$$\sin^2\beta = -(m+1), \quad \cos^2\beta = m+2, \quad \sin^2\alpha = -\frac{m+2}{2m+1}, \quad \cos^2\alpha = \frac{3m+3}{2m+1}, \tag{A27}$$

where the solution $\sin^2\beta = -1/2$ has been omitted. Thus we obtain

$$\zeta^T = \frac{1}{\sqrt{-(2m+1)}} e^{i\theta_0} \begin{bmatrix} 0, (-m-1)\sqrt{m+2}e^{-i(\theta_{-1}-\theta_0)}, & (m+2)\sqrt{-2(m+1)}, \\ (m+2)\sqrt{m+2}e^{i(\theta_{-1}-\theta_0)}, & -\sqrt{-3(m+1)}e^{2i(\theta_{-1}-\theta_0)} \end{bmatrix} \tag{A28}$$

for $-2 < m < -1$, where the range of m can be gotten from Eq. (A27) ($\sin^2\alpha, \cos^2\alpha, \sin^2\beta, \cos^2\beta > 0$).

3. For $\zeta_{-2} = 0$, in the same way, we have

$$\zeta^T = \frac{1}{\sqrt{(2m-1)}} e^{i\theta_0} \begin{bmatrix} -\sqrt{3(m-1)}e^{2i(\theta_1-\theta_0)}, & (2-m)\sqrt{2-m}e^{i(\theta_1-\theta_0)}, \\ (2-m)\sqrt{2(m-1)}, & (m-1)\sqrt{2-m}e^{-i(\theta_1-\theta_0)}, 0 \end{bmatrix} \tag{A29}$$

for $1 < m < 2$.

4. For $\zeta_1 = 0$, Eq. (A1) reduces to

$$\sqrt{6}|\zeta_0| = -2|\zeta_{-2}|e^{i(\theta_{-2}+\theta_0-2\theta_{-1})}, \tag{A30}$$

namely, $\theta_{-2} + \theta_0 - 2\theta_{-1} = \pi$, and

$$\sqrt{6}|\zeta_0| = 2|\zeta_{-2}|. \tag{A31}$$

We also get $\theta_{-2} - \theta_0 = \pi + 2(\theta_{-1} - \theta_0)$.

Equation (A2) reduces to

$$|\zeta_0|^2 = -2|\zeta_2||\zeta_{-2}|e^{i(\theta_2+\theta_{-2}-2\theta_0)}, \tag{A32}$$

namely, $\theta_2 + \theta_{-2} - 2\theta_0 = \pi$, and

$$|\zeta_0|^2 = 2|\zeta_2||\zeta_{-2}|. \tag{A33}$$

We also have $\theta_2 - \theta_0 = \pi - (\theta_{-2} - \theta_0) = -2(\theta_{-1} - \theta_0)$.

Equations (A31) and (A33) yield

$$|\zeta_{-2}| = 3|\zeta_2|, \; |\zeta_0| = \sqrt{6}|\zeta_2|. \tag{A34}$$

Substituting into Eq. (A4) gives

$$16|\zeta_2|^2 + |\zeta_{-1}|^2 = -m, \tag{A35}$$

but substituting into Eq. (A3) gives

$$16|\zeta_2|^2 + |\zeta_{-1}|^2 = 1, \tag{A36}$$

and thus $m = -1$. We take

$$|\zeta_{-1}| = \cos\alpha, \; |\zeta_2| = \tfrac{1}{4}\sin\alpha, \tag{A37}$$

where $\sin\alpha$, $\cos\alpha$ are nonzero. Then, from Eq. (A34), we obtain

$$|\zeta_0| = \frac{\sqrt{6}}{4}\sin\alpha, \; |\zeta_{-2}| = \frac{3}{4}\sin\alpha. \tag{A38}$$

Then, for $m = -1$, we have

$$\zeta^T = \tfrac{1}{4}e^{i\theta_0}[\sin\alpha e^{-2i(\theta_{-1}-\theta_0)}, 0, \sqrt{6}\sin\alpha, 4\cos\alpha e^{i(\theta_{-1}-\theta_0)}, \; -3\sin\alpha e^{2i(\theta_{-1}-\theta_0)}]. \tag{A39}$$

5. For $\zeta_{-1} = 0$, similarly, we get

$$\zeta^T = \tfrac{1}{4}e^{i\theta_0}[-3\sin\alpha e^{2i(\theta_1-\theta_0)}, 4\cos\alpha e^{i(\theta_1-\theta_0)}, \; \times\sqrt{6}\sin\alpha, 0, \; \sin\alpha e^{-2i(\theta_1-\theta_0)}] \tag{A40}$$

for $m = 1$.

E. Atoms populate at all hyperfine sublevels

Equation (A2) reduces to

$$|\zeta_0|^2 = 2|\zeta_1||\zeta_{-1}|e^{i(\theta_1+\theta_{-1}-2\theta_0)} - 2|\zeta_2||\zeta_{-2}|e^{i(\theta_2+\theta_{-2}-2\theta_0)}, \tag{A41}$$

and we can only deal with the following three special cases:

$$1.\; \theta_1 + \theta_{-1} - 2\theta_0 = 0, \; \theta_2 + \theta_{-2} - 2\theta_0 = \pi, \tag{A42}$$

$$2.\; \theta_1 + \theta_{-1} - 2\theta_0 = 0, \; \theta_2 + \theta_{-2} - 2\theta_0 = 0, \tag{A43}$$

$$3.\; \theta_1 + \theta_{-1} - 2\theta_0 = \pi, \; \theta_2 + \theta_{-2} - 2\theta_0 = \pi. \tag{A44}$$

All give the same results, and thus we take case 1 as an example. Then we have

$$|\zeta_0|^2 = 2|\zeta_1||\zeta_{-1}| + 2|\zeta_2||\zeta_{-2}|. \tag{A45}$$

We also get $\theta_{-1} - \theta_0 = -(\theta_1 - \theta_0)$, $\theta_{-2} - \theta_0 = \pi - (\theta_2 - \theta_0)$, and $\theta_{-2} - \theta_{-1} = \pi + (\theta_1 - \theta_2)$.

Equation (A1) reduces to

$$\sqrt{6}|\zeta_0|(|\zeta_1| + |\zeta_{-1}|)e^{i(\theta_0-\theta_1)} = 2(|\zeta_{-1}||\zeta_{-2}| - |\zeta_2||\zeta_1|)e^{i(\theta_1-\theta_2)}, \tag{A46}$$

and Eq. (A3) reduces to

$$(|\zeta_2| + |\zeta_{-2}|)^2 + (|\zeta_1| + |\zeta_{-1}|)^2 = 1. \tag{A47}$$

Thus we take

$$|\zeta_2| + |\zeta_{-2}| = \sin\alpha, \; |\zeta_1| + |\zeta_{-1}| = \cos\alpha, \tag{A48}$$

where $\sin\alpha > 0$, $\cos\alpha > 0$. Substituting into Eq. (A4) gives

$$\frac{2\sin\alpha}{m}(|\zeta_2| - |\zeta_{-2}|) + \frac{\cos\alpha}{m}(|\zeta_1| - |\zeta_{-1}|) = 1, \tag{A49}$$

and thus we have

$$|\zeta_2| - |\zeta_{-2}| = \frac{m}{2}\sin\alpha \;, |\zeta_1| - |\zeta_{-1}| = m\cos\alpha. \tag{A50}$$

From Eqs. (A48) and (A50), we obtain

$$|\zeta_2| = \tfrac{1}{4}(m+2)\sin\alpha, \; |\zeta_{-2}| = \tfrac{1}{4}(2-m)\sin\alpha, \\ |\zeta_1| = \tfrac{1}{2}(m+1)\cos\alpha, \; |\zeta_{-1}| = \tfrac{1}{2}(1-m)\cos\alpha, \tag{A51}$$

$$|\zeta_0|^2 = \tfrac{1}{2}(1-m^2)\cos^2\alpha + \tfrac{1}{8}(4-m^2)\sin^2\alpha. \tag{A52}$$

Substituting Eq. (A51) into Eq. (A46) yields

$$\sqrt{6}|\zeta_0| = -\tfrac{3}{2}m\sin\alpha e^{i(2\theta_1-\theta_2-\theta_0)}. \tag{A53}$$

Thus, for $m > 0$, we have $2\theta_1 - \theta_2 - \theta_0 = \pi$, namely, $\theta_2 - \theta_0 = -\pi + 2(\theta_1 - \theta_0)$ and for $m < 0$, we have $2\theta_1 - \theta_2 - \theta_0 = 0$, namely, $\theta_2 - \theta_0 = 2(\theta_1 - \theta_0)$. Then it seems that we have obtained the general form of the cyclic states:

$$\zeta^T = \frac{1}{4}e^{i\theta_0}\left[\begin{array}{c} -(m+2)\sin\alpha e^{2i(\theta_1-\theta_0)}, 2(m+1)\cos\alpha e^{i(\theta_1-\theta_0)}, \\ \sqrt{8(1-m^2)\cos^2\alpha + 2(4-m^2)\sin^2\alpha}, 2(1-m)\cos\alpha e^{-i(\theta_1-\theta_0)}, (2-m)\sin\alpha e^{-2i(\theta_1-\theta_0)} \end{array}\right] \tag{A54}$$

for $m > 0$ and

$$\zeta^T = \frac{1}{4}e^{i\theta_0}\left[\begin{array}{c} (m+2)\sin\alpha e^{-2i(\theta_{-1}-\theta_0)}, 2(m+1)\cos\alpha e^{-i(\theta_{-1}-\theta_0)}, \\ \sqrt{8(1-m^2)\cos^2\alpha + 2(4-m^2)\sin^2\alpha}, 2(1-m)\cos\alpha e^{i(\theta_{-1}-\theta_0)}, -(2-m)\sin\alpha e^{2i(\theta_{-1}-\theta_0)} \end{array}\right], \tag{A55}$$

for $m < 0$.

But we have

$$\sqrt{6}|\zeta_0| = \pm\tfrac{3}{2}m\sin\alpha, \tag{A56}$$

and Eq. (A52) then yields

$$4(1-m^2) = 0\sin^2\alpha, \tag{A57}$$

namely, $m = \pm 1$. Thus our general solutions reduce to Eqs. (A40) and (A39) for $m = 1$ and $m = -1$, respectively.

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