Diagnostics for the ground state phase of a spin-2 Bose-Einstein condensate

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We propose a method to determine the singlet-pair energy of a spin-2 Bose-Einstein condensate (BEC). By preparing the initial populations in the magnetic sublevels 0 and ±2 with appropriate relative phases, we can obtain the coefficient of the spin singlet-pair term from the spin exchange dynamics. This method is suitable for hyperfine states with short lifetimes, since only the initial change in the population of each magnetic sublevel is needed. This method therefore enables the determination of the ground state phase of a spin-2 $^{87}$Rb BEC at zero magnetic field, which is considered to lie in the immediate vicinity of the boundary between the antiferromagnetic and cyclic phases. We also show that the initial state in which relative phases are controlled can be prepared by Raman processes.

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I. INTRODUCTION

A Bose-Einstein condensate (BEC) in an optical trap [1] exhibits a rich variety of spin-related phenomena, such as various magnetic phases [2] and spin domain formation [3]. The nuclear spin of $^{87}$Rb and $^{23}$Na is 3/2, and combined with the spin 1/2 of the outermost electron, the possible hyperfine spins of these atomic species are $f = 1$ and $f = 2$. The $f = 1$ BEC was first realized at MIT with $^{23}$Na [1], for which the antiferromagnetic behavior was observed [2]. On the other hand, $^{87}$Rb in the $f = 1$ hyperfine state was found to be ferromagnetic [4,5]. The $f = 2$ $^{23}$Na condensate was also realized by the MIT group [6], whose ground state phase at zero magnetic field is predicted to be antiferromagnetic [7]. However, the ground state phase and the spin dynamics of the $f = 2$ $^{23}$Na BEC have not been studied because of its very short lifetime (a few milliseconds), due to the fact that the energy of the $f = 2$ state is higher than that of the $f = 1$ state. The $f = 2$ $^{87}$Rb BEC also lies energetically higher than its $f = 1$ counterpart, but its lifetime is much larger (~100 ms) due to a fortuitous coincidence of the singlet and triplet scattering lengths [8]. The coherent spin dynamics of the $f = 2$ BEC have been observed [9,10].

The ground state of the $f = 2$ BEC of $^{87}$Rb is predicted to be very close to the phase boundary between the antiferromagnetic and cyclic phases [7–9]. According to calculations performed by Klausen et al. [10], the ground state phase of this BEC at zero magnetic field is barely in the antiferromagnetic phase. Recent experiments performed by the Hamburg group [11] and by the Gakushuin group [9] appear to support this prediction. However, due to the experimental uncertainties, the possibility that the ground state phase is cyclic has not been excluded. In the Hamburg experiment [11], the $m = \pm 2$ mixture was shown to be stable, which is the ground state of the antiferromagnetic phase. However, a magnetic field of 340 mG was applied to the system in this experiment. Since the magnetic field lowers the energy of the $m = \pm 2$ states due to the quadratic Zeeman effect, the observed stability of the $m = \pm 2$ mixture may be due to the presence of the magnetic field. That there is no spatial separation between the $m = \pm 2$ states is a necessary condition for a BEC to be antiferromagnetic, but it is not sufficient. In the Gakushuin experiment [9], the spin dynamics starting from the $m = 0$ state were investigated for various values of the magnetic field. It was found that the population of the $m = \pm 2$ components after 70 ms evolution increased with a decrease of the magnetic field, which might imply that the $f = 2$ BEC is antiferromagnetic at zero magnetic field. However, the initial $m = 0$ state is a highly excited state and the resultant state after 70 ms is far above the ground state. Thus, the experimental observations do not exclude the possibility that the ground state phase at zero magnetic field is cyclic. In fact, the Hamburg group observed that the spin configuration of the cyclic phase has a long lifetime [11,12].

There are two experimental difficulties that hinder the determination of the ground state phase. One is the short lifetime of ~100 ms of the upper hyperfine manifold of $^{87}$Rb, which makes the equilibrium spin state hard to reach. In order to realize the ground state phase, we must equilibrate the system at least for a few seconds, as in the $f = 1$ case [2,3]. The other difficulty is the small energy scale (~0.1 nK) of the spin-singlet-pair term whose sign determines the ground state phase. In order to determine the sign of the spin-singlet pair energy, the magnetic field must be reduced so that the quadratic Zeeman energy is less than the spin-singlet pair energy. However, in such a low magnetic field the system may be affected by stray ac magnetic fields [3].

To circumvent these difficulties, we propose a method to determine the value of the spin-singlet pair energy by spin exchange dynamics. We show that by controlling the initial population and phase in each magnetic sublevel, we can measure the value of the singlet-pair energy from the initial spin dynamics. This method therefore has the advantage that equilibrating the system, which takes a

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long time, is not necessary. Furthermore, the quadratic Zeeman energy is allowed to exceed the singlet-pair energy and extreme suppression of stray magnetic fields is not required.

This paper is organized as follows. Section II formulates the mean field and Bogoliubov theories of a spin-2 BEC in the presence of the quadratic Zeeman effect. Section III studies the spin dynamics in a homogeneous system and proposes a method to determine the spin-singlet pair energy from the spin exchange dynamics. Section IV numerically verifies the proposed method for a trapped system with two-body loss. Section V discusses the initial spin preparation and Sec. VI concludes the paper. Detailed derivations of the analytic solutions are given in the Appendix.

II. MEAN FIELD AND BOGOLIUBOV ANALYSIS FOR A HOMOGENEOUS SYSTEM

A. Formulation of the problem

From rotational symmetry, the low-energy interaction between two atoms with hyperfine spin \( f \) can be classified in terms of the total spin \( F = 0, 2, \cdots, 2f \), and each scattering channel can be described by the corresponding \( s \)-wave scattering length \( a_F \). In the case of \( f = 2 \) there are three scattering channels \( F = 0, 2, 4 \), and the interaction is described by \((4\pi\hbar^2/M)(a_0p_0 + a_2p_2 + a_4p_4)\delta(r_1 - r_2)\), where \( M \) is the mass of the atom and \( p_F \) projects the state of two colliding atoms into the state of total spin \( F \). The interaction Hamiltonian can be rewritten as \((c_0 + c_1S_1 \cdot S_2 + c_2p_0)\delta(r_1 - r_2)\), where \( S_1 \) and \( S_2 \) are the spin operators for atoms 1 and 2 and the interaction coefficients are given by

\[
\begin{align*}
c_0 & = \frac{4\pi\hbar^2}{M} \left( 4a_2 + 3a_4 \right) / 7, \\
c_1 & = \frac{4\pi\hbar^2}{M} \left( 4a_4 - 3a_2 \right) / 7, \\
c_2 & = \frac{4\pi\hbar^2}{M} \left( 7a_0 - 10a_2 + 3a_4 \right) / 7.
\end{align*}
\]

For the linear and quadratic Zeeman effects, the energy of the atom depends on the magnetic field \( B \) as \( pm + qm^2 \), where \( p = \mu_B B/2 \pi \) and \( q = -(\mu_B B^2)/(4\Delta_{hf}) \) with \( \mu_B \) the Bohr magneton and \( \Delta_{hf} \) the hyperfine splitting between the states \( f = 1 \) and \( f = 2 \). We assume \( q < 0 \) throughout this paper, which is the case for spin-2 \(^{87}\text{Rb}\).

The mean-field energy of the entire system is given by

\[
E = \int dr \left\{ \sum_{m=-2}^{2} |\psi_m|^2 \left( -\frac{\hbar^2}{2M} \nabla^2 + V + pm + qm^2 - \frac{c_0}{2} n_{tot}^2 + \frac{c_1}{2} F^2 + \frac{c_2}{2} |A_{00}|^2 \right) \psi_m \right\},
\]

where \( m = -2, \cdots, 2 \) denotes the magnetic sublevels, \( V \) is an external potential,

\[
n_{tot} = \sum_{m=-2}^{2} |\psi_m|^2
\]

is the total particle-number density,

\[
F = \sum_{mm'} \psi_m^* S_{mm'} \psi_{m'}
\]

is the spin vector with \( 5 \times 5 \) spin-2 matrix \( S \), and

\[
A_{00} = \frac{1}{\sqrt{5}} \left( 2\psi_2 \psi_{-2} - 2\psi_1 \psi_{-1} + \psi_0^2 \right)
\]

is the singlet-pair amplitude. Minimizing the action,

\[
S = \int dt (ih \sum_m \psi_m^* \partial_t \psi_m - E),
\]

gives the multicomponent Gross-Pitaevskii (GP) equations for the spin-2 BEC as

\[

i\hbar \frac{\partial \psi_{\pm 2}}{\partial t} = \left( -\frac{\hbar^2}{2M} \nabla^2 + V \mp 2p + 4q \right) \psi_{\pm 2} + c_0 n_{tot} \psi_{\pm 2}
\]

\[
+ c_1 \left( F_+ \psi_{\pm 1} \mp 2F_z \psi_{\pm 2} \right) + \frac{c_2}{\sqrt{5}} A_{00} \psi_{\mp 2}^2,
\]

\[
\frac{\hbar}{2M} \nabla^2 + V \right) \psi_{\pm 1} + c_0 n_{tot} \psi_{\pm 1}
\]

\[
+ c_1 \left( \frac{\sqrt{6}}{2} F_+ \psi_0 + F_\mp \psi_{\pm 2} \pm F_z \psi_{\pm 1} \right)
\]

\[
- \frac{c_2}{\sqrt{5}} A_{00} \psi_{\mp 1}^*,
\]

\[
\frac{\hbar}{2M} \nabla^2 + V \right) \psi_0 + c_0 n_{tot} \psi_0
\]

\[
+ \frac{\sqrt{6}}{2} c_1 \left( F_- \psi_{-1} + F_+ \psi_1 \right) + \frac{c_2}{\sqrt{5}} A_{00} \psi_0^*,
\]

where \( F_\pm = F_\pm \pm iF_\mp \).

We note that the linear Zeeman term only rotates the hyperfine spin about the \( z \) axis, and hence the spin dynamics are essentially independent of the linear Zeeman term. In fact, setting \( \psi_m \rightarrow e^{ipmt/\hbar} \psi_m \) and noting that the phase factor also arises in the \( F_\pm \) terms as \( e^{i\psi F_\pm t/\hbar} \), we can completely eliminate the linear Zeeman terms in Eqs. (7a) and (7b). This property is due to the rotational symmetry of our system with respect to the \( z \) axis, which results in the conservation of the projected angular momentum on the \( z \) axis, \( \langle F_z \rangle = \int d\mathbf{r} \sum_m m |\psi_m|^2 \).

B. Ground states in a homogeneous system

In an ultracold spinor BEC which is isolated from the environment, the total spin angular momentum in the direction of the magnetic field is conserved for a long time.
We therefore minimize the energy of the system in the subspace of a given \( \langle F_2 \rangle = \int d\mathbf{r} \sum_m m |\psi_m|^2 \); we shall refer to the resulting minimized state as the “ground state”. For simplicity, we restrict ourselves to the subspace of \( \langle F_2 \rangle = 0 \) in this paper, i.e., we seek the ground state in the subspace of \( \langle F_2 \rangle = 0 \).

We consider the uniform case by setting \( V = 0 \) and \( \psi_m = \sqrt{N} \zeta_m \), where the density \( n \) is constant and \( \zeta_m \) satisfies \( \sum_m |\zeta_m|^2 = 1 \). The energy per atom is given by

\[
\frac{\varepsilon}{N} = q \sum_{m=-2}^{2} m^2 |\zeta_m|^2 + \frac{\tilde{c}_0}{2} f^2 + \frac{\tilde{c}_2}{2} |a_{00}|^2,
\]

where \( f = F/n = \sum_{m \neq 0} \zeta_m^* S_{mm'} \zeta_{m'} \), \( a_{00} = A_{00}/n = (2\zeta_2 - 2\zeta_1 + \zeta_0^2)/\sqrt{3} \), and

\[
\tilde{c}_i \equiv c_i n
\]

for \( i = 0, 1, 2 \). The linear Zeeman term is absent in Eq. 8 because \( \langle F_2 \rangle = 0 \).

In order to find the ground state, we compare the energies of the stationary states given in Ref. 11. The energy of the antiferromagnetic state,

\[
\zeta_{AF} = (e^{ixz}, 0, 0, 0, e^{ix-z}),
\]

where \( \chi_m \) is the phase of each component, is given by

\[
\varepsilon_{AF} = 4q + \tilde{c}_0/2 + \tilde{c}_2/10.
\]

This energy is independent of the phase factor \( e^{ixm} \). For the cyclic state,

\[
\zeta_C = \left( \frac{1}{\sqrt{2}} e^{ixz} \sin \theta, 0, e^{ix0} \cos \theta, 0, \frac{1}{\sqrt{2}} e^{ix-z} \sin \theta \right),
\]

the energy of the system takes the form

\[
\varepsilon_C = \frac{\tilde{c}_0}{2} + \frac{\tilde{c}_2}{10} \left[ \cos^2 \theta + e^{ix} \sin^2 \theta \right] + 4q \sin^2 \theta,
\]

where \( \chi \equiv \chi_2 + \chi-z - 2\chi_0 \). When \( \tilde{c}_2 < 10|q| \), Eq. 13 is minimized by \( \theta = \pi/k_B \), which corresponds to the antiferromagnetic state \( \zeta_{AF} \). When \( \tilde{c}_2 > 10|q| \), Eq. 13 becomes minimal with \( \chi = \pi \) and

\[
\cos^2 \theta = \frac{1}{2} \frac{5q}{\tilde{c}_2}.
\]

Substituting this into Eq. 13, we obtain

\[
\varepsilon_C = 2q + \frac{\tilde{c}_0}{2} - \frac{10q^2}{\tilde{c}_2} \quad (\tilde{c}_2 > 10|q|).
\]

The states \( \zeta_{AF} \) and \( \zeta_C \) satisfy \( F_z(r) = \sum_m m |\psi_m(r)|^2 = 0 \).

Even if the local spin \( F_z(r) \) is nonzero, the total spin \( \langle F_z \rangle \) can be zero by the formation of the spatial spin structure. For example, a staggered domain structure of the ferromagnetic state gives \( \langle F_z \rangle = 0 \). The kinetic energy at the domain wall can be neglected when the region of the wall is much smaller than the region of the domain. Hence, we also consider the case of \( F_z(r) \neq 0 \), bearing in mind that the total spin is maintained as zero, \( \langle F_z \rangle = 0 \), by some structure formation. The energy of the ferromagnetic state, \( \zeta_{F} = (e^{ix2}, 0, 0, 0, 0) \) or \( (0, e^{ix1} \sin \theta, 0, 0, e^{ix-z} \cos \theta) \), is given by

\[
\varepsilon_F = 4q + \tilde{c}_0/2 + 2\tilde{c}_1.
\]

We also consider a state found in Ref. 11,

\[
\zeta_M = (e^{ix2} \cos \theta, 0, 0, e^{ix-1} \sin \theta, 0) \text{ or } (0, e^{ix1} \sin \theta, 0, 0, e^{ix-z} \cos \theta).
\]

The energy of this state is obtained to be

\[
\varepsilon_M = q(1 + 4 \cos^2 \theta) + \frac{\tilde{c}_0}{2} + \frac{\tilde{c}_1}{2} (3 \cos^2 \theta - 1)^2.
\]

This is minimized by \( \cos^2 \theta = 1/3 - q/(3\tilde{c}_1) \) for \( \tilde{c}_1 > |q|/2 \), and the energy then becomes

\[
\varepsilon_M = 2q + \frac{\tilde{c}_0}{2} - \frac{q^2}{2\tilde{c}_1} \quad (\tilde{c}_1 > |q|/2).
\]

Comparing the energies \( \varepsilon_{AF}, \varepsilon_F, \varepsilon_C, \) and \( \varepsilon_M \), we obtain the phase diagrams shown in Fig. 1. We have assumed here that one of the states \( \zeta_{AF}, \zeta_C, \zeta_F, \) and \( \zeta_M \) is the ground state, based on the numerical results of Ref. 11. We have confirmed that this assumption is correct using the Monte Carlo method.

In the case of \( ^{87}\text{Rb} \), \( \tilde{c}_1 \) is positive and \( \tilde{c}_2 \) is at most of the same order of magnitude as \( \tilde{c}_1 \). Hence, the ground state of the spin-2 \( ^{87}\text{Rb} \) BEC is antiferromagnetic or cyclic depending on whether \( \tilde{c}_2 < 10|q| \) or \( \tilde{c}_2 > 10|q| \). Therefore, in order to determine the sign of \( \tilde{c}_2 \) from the ground state, we must suppress the magnetic field so that the condition \( |q| < \tilde{c}_2/10 \) is met. In the experiment of the Hamburg group, a magnetic field of \( B = 340 \text{ mG} \) was applied, which corresponds to \( |q|/k_B \approx 0.4 \text{ nK} \). The mean atomic density was \( n \approx 4 \times 10^{14} \text{cm}^{-3} \), and when \( |\tilde{c}_2|/k_B \approx 0.8 \text{ nK} \), we have \( |\tilde{c}_2|/k_B \approx 1.5 \text{ nK} \). Thus, \( \tilde{c}_2 \approx 10|q| \), which is consistent with the observed stability of the antiferromagnetic state \( \zeta_{AF} \). In the Gakushuin experiment, \( B > 100 \text{ mG} \) and \( n \approx 2.1 \times 10^{14} \text{cm}^{-3} \), corresponding to \( |q| \approx 0.03 \text{ nK} \) and \( |\tilde{c}_2|/k_B \approx 0.8 \text{ nK} \). Hence, the reported parameters of Ref. 12 belong to the cyclic phase.

### C. Stability of the stationary states

The antiferromagnetic (or stretched) state, 

\[
\Psi_{AF} = \sqrt{2}(1, 0, 0, 0, 1),
\]

was found in experiments to be stable in the presence of a magnetic field. To examine this result, we will
investigate the stability of this state using Bogoliubov analysis.

Substituting Eq. (20) into Eqs. (7), we find that it evolves as
\[ e^{-i\mu t/\hbar} (e^{-2i\pi t/\hbar} 0, 0, e^{2i\pi t/\hbar}) \text{ with } \mu = 4q + \tilde{c}_0 + \tilde{c}_2/5, \] and hence Eq. (20) is stationary in the rotating frame \( e^{i\mu t/\hbar} \). We can therefore perform Bogoliubov analysis with respect to the state \( |\Psi_{\text{AF}}\rangle \) in the rotating frame, which is described by the GP equations (7) without the linear Zeeman terms. We set the wave function as \( \psi = e^{-i\mu t/\hbar} (\Psi_{\text{AF}} + \phi) \) and substitute it into the GP equations in the rotating frame. Taking the linear terms with respect to \( \phi \), we obtain

\[
\frac{i\hbar}{\partial t} \phi_{\pm 2} = -\frac{\hbar^2}{2M} \nabla^2 \phi_{\pm 2} + \left( \frac{\tilde{c}_0}{2} + 2\tilde{c}_1 \right) (\phi_{\pm 2} + \phi_{\pm 2}^*),
\]

\[
\frac{i\hbar}{\partial t} \phi_{\pm 1} = -\frac{\hbar^2}{2M} \nabla^2 - 3q \phi_{\pm 1} + \left( \frac{\tilde{c}_1 - \tilde{c}_2}{5} \right) \phi_{\pm 1} + \phi_{\pm 1}^*,
\]

\[
\frac{i\hbar}{\partial t} \phi_0 = -\frac{\hbar^2}{2M} \nabla^2 - 4q \phi_0 + \frac{\tilde{c}_2}{5} \phi_0 + \frac{\tilde{c}_2}{5} \phi_0^*.
\]

These equations can be reduced to the eigenvalue problem by expansion of \( \phi \) as

\[
\phi(r, t) = \sum_k \left( u_k e^{i(k\cdot r - \omega_k t)} + v_k e^{-i(k\cdot r - \omega_k t)} \right),
\]

and the eigenenergies are obtained as

\[
[\tilde{\varepsilon}_k (\tilde{\varepsilon}_k + 2\tilde{c}_0 + 2\tilde{c}_2/5)]^{1/2},
\]

\[
[\tilde{\varepsilon}_k (\tilde{\varepsilon}_k + 8\tilde{c}_1 - 2\tilde{c}_2/5)]^{1/2},
\]

\[
[(\tilde{\varepsilon}_k - 3q) (\tilde{\varepsilon}_k - 3q + 2\tilde{c}_1 - 2\tilde{c}_2/5)]^{1/2},
\]

\[
[(\tilde{\varepsilon}_k - 4q) (\tilde{\varepsilon}_k - 4q - 2\tilde{c}_2/5)]^{1/2},
\]

where \( \tilde{\varepsilon}_k \equiv (\hbar k)^2/(2M) \).

If the eigenenergy is complex, the corresponding mode is dynamically unstable against exponential growth. The eigenvectors \( u_k \) and \( v_k \) of the first mode (23a) are proportional to \((1, 0, 0, 0, 1)\), which is the same form as \( \Psi_{\text{AF}} \), indicating that a collapse occurs if \( 2\tilde{c}_0 + 2\tilde{c}_2/5 < 0 \). The eigenvectors of the second mode (23b) are proportional to \((1, 0, 0, 0, -1)\). Since this eigenmode transfers the \( m = \pm 2 \) component to the \( m = \mp 2 \) component, excitation of the mode (23b) gives rise to exchange of atoms between the \( m = \pm 2 \) components. This implies that phase separation between the two components occurs for \( 8\tilde{c}_1 - 2\tilde{c}_2/5 < 0 \). Therefore, the fact that no phase separation is observed in experiments [4] only indicates that \( \tilde{c}_2 < 20\tilde{c}_1 \); it is not sufficient to conclude that \( \tilde{c}_2 < 0 \). The third mode (23c) is two-fold degenerate and the eigenvectors are proportional to \((0, 1, 0, 0, 0)\) and \((0, 0, 1, 0, 0)\). Therefore, when \( 2\tilde{c}_1 - 2\tilde{c}_2/5 < 3q \), the state (20) is dynamically unstable against the growth of the \( m = \pm 1 \) components. Similarly the last mode (23d) is proportional to \((0, 0, 1, 0, 0)\) and describes the growth of the \( m = 0 \) component. Dynamical instability arises in this mode for \( \tilde{c}_2 > 10|q| \), consistent with the fact that the ground state has the \( m = 0 \) component for \( \tilde{c}_2 > 10|q| \), as shown by Eq. (13).

For later use, we also perform Bogoliubov analysis for the stationary state \( \Psi = (0, 0, \sqrt{n}, 0, 0) \). The eigener-
The spin independent interaction does not flip the spin. The eigenvectors of the mode \(24a\) are proportional to \((0, 0, 1, 0, 0) \propto \Psi\), implying that the state \(\Psi\) collapses if \(2\tilde{c}_0 + 2\tilde{c}_2/5 < 0\). We note that this condition for the collapse of the BEC is the same as that for \(\Psi_{AF}\). The second and third modes, Eqs. \(24b\) and \(24c\), are both two-fold degenerate. The eigenvectors of the mode \(24c\) are proportional to \((0,1,0,0,0)\) and \((0,0,0,1,0)\), and therefore the dynamical instability in this mode increases the \(m = \pm 1\) components. Similarly, the mode \(24c\) has eigenvectors proportional to \((1,0,0,0,0)\) and \((0,0,0,0,1)\). In contrast to the excitations for \(\Psi_{AF}\), there are always dynamically unstable modes in Eqs. \(24b\) and \(24c\) because of \(q < 0\), and therefore the \(m = 0\) state is always dynamically unstable for \(B > 0\) in a homogeneous system.

III. SPIN DYNAMICS IN A HOMOGENEOUS SYSTEM

A. Analytic solutions

In order to determine the value of \(c_2\) from the spin dynamics, we consider the spin dependent interaction that is sensitive only to the spin-singlet term. The elementary processes in the spin-singlet channel are \(0 + 0 \leftrightarrow 0 + 0\), \(0 + 0 \leftrightarrow 1 + (-1)\), \(0 + 0 \leftrightarrow 2 + (-2)\), and \(1 + (-1) \leftrightarrow 2 + (-2)\). On the other hand, the \(c_1\) term consists of elementary processes such as \(m_1 + m_2 \leftrightarrow (m_1 + 1) + (-m_2 - 1)\). The spin independent interaction does not flip the spin. Thus, the elementary process appearing only in the \(c_2\) term is \(0 + 0 \leftrightarrow 2 + (-2)\). We therefore focus on this process and consider the initial state of the form

\[
\zeta = \left( \frac{1}{\sqrt{2}} e^{i\chi_2} \sin \theta, 0, e^{i\chi_0} \cos \theta, 0, \frac{1}{\sqrt{2}} e^{i\chi_2 - \sin \theta} \right). \tag{25}
\]

Since the overall phase and spin rotation about the \(z\) axis do not affect the physics, the relevant phase appears only in the combination of \(\chi_2 + \chi_2 - 2\chi_0\). We therefore assume that \(\chi_0 = \chi_2 = 0\) and \(0 \leq \theta \leq \pi/2\) without loss of generality.

If \(\zeta_{\pm 1}\) are exactly zero in the initial state, we find from Eq. \(10\) that \(\zeta_{\pm 1}(t)\) are always zero within the mean field approximation. We therefore assume \(\zeta_{+1}(t) = 0\) in the following analysis. In experiments, this assumption holds at least for an initial period of \(\sim 100\) ms. Although the \(m = \pm 1\) components exponentially grow in the presence of the dynamical instability, the assumption \(\zeta_{\pm 1}(t) = 0\) is still valid in the early stage of time evolution.

The GP equations then reduce to

\[
\begin{align*}
\hbar \frac{d}{dt} \zeta_0 &= \frac{\tilde{c}_2}{5} \zeta_0^* (2\zeta_2 \zeta_2 + \zeta_0^2), \tag{26a} \\
\hbar \frac{d}{dt} \zeta_{\pm 2} &= \frac{\tilde{c}_2}{5} \zeta_{\mp 2}^* (2\zeta_2 \zeta_2 + \zeta_0^2) + 4q \zeta_{\pm 2}. \tag{26b}
\end{align*}
\]

Differentiating Eq. \(26a\) with respect to time and using Eq. \(25\), we can eliminate \(\zeta_{\pm 2}\) to obtain

\[
h^2 \zeta_0 = \frac{2\tilde{c}_2}{5} (\zeta_0^2 - 4q |\zeta_0|^2) \zeta_0 - i \left( \frac{2\tilde{c}_2}{5} + 8q \right) \hbar \zeta_0, \tag{27}
\]

where the energy per atom,

\[
\varepsilon = \frac{\tilde{c}_2}{10} (2\zeta_2 \zeta_2 + \zeta_0^2)^2 + 4q (|\zeta_2|^2 + |\zeta_2|^2), \tag{28}
\]

is a constant of motion. It is interesting to note that the form of Eq. \(27\) coincides with that describing a 1D BEC on a rotating ring if the time derivative is replaced with the spatial derivative [17]. The solution is obtained (see Appendix for derivation) as

\[
\zeta_0(t) = e^{i\varphi_0(t)} \sqrt{A_0} \sin^2 (\alpha t + \beta_0 \nu) + B_0, \tag{29}
\]

where \(\varphi_0(t), A_0, B_0, \alpha, \beta_0, \) and \(\nu\) are given by Eqs. \(A24\), \(A10\), \(A15\), \(A21\), \(A22\), and \(A17\), respectively. The function \(\sin^2\) is a Jacobian elliptic function [18].

If we assume \(\chi_2 = \pi\) in the initial state, i.e.,

\[
\zeta(0) = \left( \frac{1}{\sqrt{2}} \sin \theta_0, 0, \cos \theta_0, 0, -\frac{1}{\sqrt{2}} \sin \theta_0 \right), \tag{30}
\]

the constants in the solution \(29\) become \(\beta_0 = 0\), \(B_0 = \zeta_{\varphi_0}^2(0) = \cos^2 \theta_0\), and

\[
A_0 = \frac{\tilde{c}_2}{10q} \zeta_{\varphi_0}^2(0) \left[ 1 - \zeta_{\varphi_0}^2(0) \right], \tag{31}
\]

\[
\alpha = \sqrt{\frac{8q}{5\hbar^2} \left( \tilde{c}_2 [1 - 2\zeta_{\varphi_0}^2(0)] + 10q \right)}, \tag{32}
\]

\[
\nu = \frac{\tilde{c}_2^2 \zeta_{\varphi_0}^2(0) \left[ 1 - \zeta_{\varphi_0}^2(0) \right]}{10q \tilde{c}_2 [1 - 2\zeta_{\varphi_0}^2(0)] + 10q}. \tag{33}
\]

The sign of \(A_0\) is opposite to that of \(c_2\) because \(q < 0\). The constants \(\alpha\) and \(\nu\) are real and positive if \(\tilde{c}_2 [1 - 2\zeta_{\varphi_0}^2(0)] + 10q < 0\).

B. Measurement of \(c_2\)

We suppose that the initial state \(30\) is prepared and the magnetic field satisfies \(|q| > |\tilde{c}_2|/10\). Under this condition, we find that \(\alpha\) and \(\nu\) in Eqs. \(32\) and \(33\) are real and positive. Since the atomic density is typically \(n \sim 10^{14} \text{cm}^{-3}\) in experiments [4, 9] and \(|(7a_0 - 10a_2 + 3a_4)/7|\) is of order of the Bohr radius at most [14], the condition \(|q| > |\tilde{c}_2|/10\) requires \(B \gtrsim 100\)
The Jacobian elliptic function $sn(\alpha t|\nu)$ initially increases for positive $\alpha$ and $\nu$, and hence the $m = 0$ component $|\zeta_0(t)|^2$ in Eq. (20) initially decreases for $c_2 > 0$ and increases for $c_2 < 0$. Therefore, we can determine the sign of $c_2$ from the sign of the initial change in the spin population, which is the main result of this paper. This method is suitable for atomic species with short lifetimes, since only the initial stage of time evolution is needed for the determination of $c_2$.

Figure 2 shows time evolution of the spin populations which are obtained by numerically solving Eq. (20). The results confirm the relation between the sign of $c_2$ and the sign of the initial change in the spin populations. In Fig. 2 the $m = \pm 1$ components are assumed to have small initial values ($\zeta_{\pm 1} = 0.001$) to assess imperfect population transfer in realistic situations. We find that the dynamical instability in the $m = \pm 1$ components have little effect on the initial dynamics of the $m = 0$, $\pm 2$ components.

In Fig. 2 $\nu \sim 0.01$. For $0 \leq \nu \ll 1$, the Jacobian elliptic function can be approximated by the trigonometric function as

$$sn(\alpha t|\nu) = \sin \alpha t + O(\nu),$$

(34)

and the spin evolution is then approximated by

$$|\zeta_0(t)|^2 \simeq \cos^2 \theta_0 + A_0 \sin^2 \alpha t,$$

(35a)

$$|\zeta_{\pm 2}(t)|^2 \simeq \frac{1}{2}(\sin^2 \theta_0 - A_0 \sin^2 \alpha t).$$

(35b)

The amplitude of this oscillation is $A_0 \propto -c_2$, showing explicitly that whether the spin population initially increases or decreases depends on the sign of $c_2$. We can also determine the magnitude of $c_2$ by measuring the amplitude of the oscillations. In Fig. 2, Eq. (35) is plotted as dashed curves, which are in good agreement with the numerical solutions (solid curves) in the early stages of the time evolution.

The differences between the numerical and analytic curves in Fig. 2 arise mainly from the growth of the $m = \pm 1$ components in the numerical result due to the dynamical instability. We find that the growth of the $m = \pm 1$ components in Fig. 2 (b) is larger than that in Fig. 2 (a). This can qualitatively be understood from the Bogoliubov energy (24b), which describes the growth of the $m = \pm 1$ components from the state $\Psi = (0,0,\sqrt{\pi},0,0)$. The imaginary part of Eq. (24b) at $\varepsilon_k = 0$, $[q(q + 6c_1 - 2c_2/5)]^{1/2}$, is larger for $c_2 < 0$ than for $c_2 > 0$ since $q < 0$ and $q + 6c_1 > 0$ in the present case.

We note that the relative phase $\chi \equiv \chi_2 + \chi_2 - 2\chi_0 = \pi$ in the initial state (25) is crucial for determining the value of $c_2$ by the above method. For example, when the initial state is given by Eq. (25) with $\chi = 0$, or without loss of generality,

$$\zeta = \left(\frac{1}{\sqrt{2}} \sin \theta_0, 0, \cos \theta_0, 0, \frac{1}{\sqrt{2}} \sin \theta_0 \right),$$

(36)

the time evolution becomes Eq. (29), where $A_0$, $\nu$, and $\alpha$ are given by Eqs. (A3), (A3), and (A3), respectively (see Appendix for details). These constants satisfy $c_2A_0 > 0$ and $\nu < 0$, and $\alpha$ is real. Using the relation

$$sn(\alpha t | -|\nu|) = \frac{1}{\sqrt{1 + |\nu|}}sd(\sqrt{1 + |\nu|} \alpha t),$$

(37)

and noting that the Jacobian elliptic function $sd = sn/dn$ initially increases, we find that $|\zeta_0(t)|^2$ initially decreases for $c_2 < 0$ and increases for $c_2 > 0$. This behavior is opposite to that in Fig. 2 and therefore control of the relative phase $\chi$ in the initial state is crucial for determining the sign of $c_2$. Figure 3 shows the initial evolution of $|\zeta_0|^2$ with $\chi$ varying from 0 to $\pi$, which indicates that the phase of the oscillation depends on $\chi$.

C. Dynamics in the cyclic phase

If $|q| < \tilde{c}_2/10$, the stretched state (20) is dynamically unstable against growth of the $m = 0$ component, as shown by Eq. (23a). This is because the ground state is in the cyclic phase given by Eq. (12) if $|q| < \tilde{c}_2/10$ and $\tilde{c}_2 < 20c_1$. We can therefore conclude that $\tilde{c}_2 - 10|q| > 0$ and hence $c_2 > 0$, if we observe the growth of the $m = 0$ component from the initial stretched state.

Suppose that $\cos^2 \theta_0 \ll 1$ in the initial state given by Eqs. (30) and $|q| < \tilde{c}_2/10$, then $\nu$ is negative and $\alpha$ is pure imaginary from Eqs. (32) and (33). In this case, the
The initial state is given by $\zeta = 0$, $\zeta = \sqrt{0.35}$, and $\zeta = \exp(i\pi/4)\sqrt{0.35}$, where $j$ is an integer ranging from 0 to 4.

Jacobian elliptic function can be rewritten as

$$\text{sn}(i|\alpha|t - |\nu|) = \frac{i}{\sqrt{1 + |\nu|}} \text{sd} \left( \sqrt{1 + |\nu|} |\alpha|t \right) \cdot \frac{1}{1 + |\nu|}. \quad (38)$$

Since $|\nu| \ll 1$ and hence $1/(1 + |\nu|) \approx 1$, we can make the approximation

$$\text{sd}(u)(1 + |\nu|)^{-1} \approx \frac{\sinh u + \frac{|\nu|}{2}(|\nu| \cosh u - u)\text{sech}}{1 + \frac{|\nu|}{2}(|\nu| \cosh u - u) \tanh u}, \quad (39)$$

where $u = |\alpha|t$. When $u \gg 1$, Eq. (39) reduces to

$$\text{sd}(u)(1 + |\nu|)^{-1} \approx \frac{1}{\sqrt{|\nu|}} \text{sech} \left( |\alpha|t - \frac{1}{2} \ln \frac{16}{|\nu|} \right). \quad (40)$$

Equation (38) is thus approximated to give

$$|\zeta_0(t)|^2 \approx -\frac{A_0}{|\nu|} \text{sech}^2 \left( |\alpha|t - \frac{1}{2} \ln \frac{16}{|\nu|} \right) + \cos^2 \theta_0. \quad (41)$$

The dashed curve in Fig. 4 shows Eq. (41). A comparison of this with the corresponding numerical result (solid curve) shows that Eq. (41) is in good agreement with the numerical result, with a small discrepancy arising mainly from the growth of the $m = \pm 1$ components in the numerical result due to the dynamical instability. The $m = 0$ component reaches a maximum at

$t \approx \ln(16/|\nu|)/(2|\alpha|)$ and the maximum value of $|\zeta_0(t)|^2$ is given by $\approx -A_0/|\nu| \approx 1 + 10q/\tilde{c}_2$.

We note that a characteristic time scale of the dynamics in Fig. 4 is $\sim 100$ ms, which is much larger than the $\sim 10$ ms seen in Fig. 2. This is because the former time scale is set by the dynamical instability, while the latter is set by the time evolution from a non-stationary state. This method can thus determine the value of $c_2$, provided that (i) $c_2$ is positive, (ii) the magnetic field is suppressed so that $|q| < \tilde{c}_2/10$, and (iii) the time scale of the dynamics is much faster than the lifetime of the $f = 2$ state. If $c_2$ is negative, the present method cannot determine the value of $c_2$.

In the case of Fig. 4 ($|q| < \tilde{c}_2/10$), the $m = 0$ population initially increases for $c_2 > 0$, which is opposite to the case of Fig. 2 ($|q| > |c_2|/10$). Nevertheless, the method to determine the sign of $c_2$ in Sec. III is still applicable, since we can easily find whether we are in a region of $|q| > \tilde{c}_2/10$ or $|q| < \tilde{c}_2/10$. If the amplitude of the initial oscillation monotonically decreases without changing sign with an increase in the magnetic field, we can conclude that $|q| > \tilde{c}_2/10$.

**IV. SPIN DYNAMICS IN THE TRAPPED SYSTEM**

We investigate the spin dynamics in a trap potential to verify the results in Sec. III in a realistic situation. We use an axisymmetric trap potential with radial and axial frequencies ($\omega_r, \omega_z$) = $2\pi \times (237, 21)$ Hz, which was used in the experiment of Ref. 3. The two-body loss rate was found to be roughly independent of the magnetic field and therefore it is considered to be insensitive to the spin-mixing dynamics 3. We therefore take into account the two-body loss by adding the term $-\hbar K_2|\psi_m|^2\psi_m/2$ to the right-hand side of Eq. 7.
FIG. 5: Time evolution of the relative population $f dr |\psi_m|^2 / N$ of each spin component $m$ in the trap with $\omega_\perp = 237$ Hz and $\omega_z = 21$ Hz. The scattering lengths are assumed to be $(c_0, c_1, c_2)M/(4\pi a_0^2) = (100a_B, a_B, a_B)$ in (a) and $(100a_B, a_B, -a_B)$ in (b), where $a_B$ is the Bohr radius. The strength of the magnetic field is 250 mG, and the two-body loss coefficient is $K_2 = 1.4 \times 10^{-13}$ cm$^3$ s$^{-1}$. The $m = -2$ ground state $\psi_0$ is prepared, and is initially transferred to each spin component as $\psi_0 = \sqrt{0.298} \psi_0$, $\psi_{\pm 1} = \sqrt{0.001} \psi_0$, and $\psi_2 = -\psi_{-2} = \sqrt{0.35} \psi_0$. The insets show the column density distributions of the $m = -2, \ldots, 2$ components from top to bottom at $t = 100$ ms, each having a $70 \times 8\mu m$ field of view.

with $K_2 = 1.4 \times 10^{-13}$ cm$^3$ s$^{-1}$. We prepare the ground state $\psi_g$ for the $m = -2$ state by the imaginary-time propagating method and transfer it to each spin component as $\psi_0 = \sqrt{0.298} \psi_0$, $\psi_{\pm 1} = \sqrt{0.001} \psi_0$, and $\psi_2 = -\psi_{-2} = \sqrt{0.35} \psi_0$. We use this state as an initial state, where the initial number of atoms is assumed to be $N(t = 0) = 4.7 \times 10^5$.

The time evolution of each spin population obtained by numerically solving the GP equation with the two-body loss terms is shown in Fig. 5. We find that the aforementioned dependence of the initial spin evolution on the sign of $c_2$ is valid in the trapped system, i.e., the quantity $|\zeta_0|^2 = f dr |\psi_0|^2 / N$ first decreases for $c_2 > 0$ and increases for $c_2 < 0$. The growth in $|\psi_{\pm 1}|^2$ due to the dynamical instability and the two-body loss does not affect this dependence. Thus, we have confirmed that the sign of $c_2$ can be determined from the spin dynamics in a realistic situation. We find that the growth of the $m = \pm 1$ components is larger for $c_2 < 0$ than for $c_2 > 0$, which is similar to the homogeneous case shown in Fig. 2.

The insets in Fig. 6 show the column density distribution of each spin component at $t = 100$ ms. The initial distribution is almost preserved at $t = 100$ ms and no spin domains are formed.

V. PREPARATION OF THE INITIAL SPIN STATE

As shown in Fig. 5, our method to determine the sign of $c_2$ hinges on the relative phase $\chi_2 + \chi_{-2} - 2\chi_0$ in the initial state. Therefore we must control the phase experimentally. In the experiments of Refs. 1, 2, the rapid-adiabatic-passage technique was employed to prepare the initial state. This method can control the phase in the initial state in principle, if the relative phases between the applied rf fields are controlled. We propose here a new method to prepare the initial state using the Raman transition.

We consider the Raman transition, for example, between the $f = 2$ and $f' = 2$ $D_1$ states by circularly polarized photons, as illustrated in Fig. 6. The initial state is assumed to be the $m = 0$ lower state. Hence, the relevant states are the $m = 0, \pm 2$ lower states and the $m = \pm 1$ upper states, whose amplitudes are denoted by $\zeta_0, \zeta_{\pm 2}$, and $\zeta_{\pm 1}$, respectively. The equations of motion...
are given by

\[ i\hbar \zeta_{\pm 2} = (\pm 2p + 4q)\zeta_{\pm 2} + \frac{1}{\sqrt{6}} g_{\mp} e^{i\omega_{\pm} t} \zeta_{\mp 1}, \tag{42a} \]
\[ i\hbar \zeta_{\pm 1} = (E_{D1} \pm p' + q')\zeta_{\pm 1} + \frac{1}{\sqrt{6}} g_\epsilon e^{-i\omega_{\pm} t} \zeta_{\pm 2} \]
\[ \pm \frac{1}{2} g_\epsilon e^{-i\omega_{\pm} t} \zeta_{0}, \tag{42b} \]
\[ i\hbar \zeta_{0} = \frac{1}{2} g_\epsilon e^{i\omega_{\pm} t} \zeta_{1} - \frac{1}{2} g_\epsilon e^{-i\omega_{\pm} t} \zeta_{-1}, \tag{42c} \]

where \( g_{\pm} \) and \( \omega_{\pm} \) are the coupling constants and frequencies of the \( \sigma_{\pm} \) photons, \( E_{D1} \) is the transition energy of the \( D_1 \) line, and \( p' \) and \( q' \) are the linear and quadratic Zeeman energies for the upper hyperfine manifold. The coupling constants \( g_{\pm} \) are given by the product of the amplitude of the circularly polarized light and the dipole matrix element between the \( s \) and \( p \) states. Using the new variables \( \zeta_{\pm 2} = e^{\pm 2i\phi_{\pm} \zeta_{\pm 2}} \) and \( \zeta_{\pm 1} = e^{i\omega_{\pm} t} \zeta_{\pm 1} \), the coefficients on the right-hand side of Eq. (42) become time independent. Assuming \( \omega_{\pm} = E_{D1} - \Delta \pm p' \) and \( \Delta \gg |p - p'| \), and eliminating \( \zeta_{\pm 1} \) from Eq. (42), we obtain

\[ i\hbar \zeta_{\pm 2} = \left( 4q - \frac{|g_{\mp}|^2}{6\Delta} \right) \zeta_{\pm 2} + i\hbar \Omega \zeta_{0}, \tag{43a} \]
\[ i\hbar \zeta_{0} = -\frac{1}{4\Delta} (|g_{\mp}|^2 + |g_{\pm}|^2) \zeta_{0} - i\hbar (\Omega^* \zeta_{2} - \Omega \zeta_{-2}), \tag{43b} \]

where \( i\hbar \Omega = g_{\mp} g_{\pm}^*/(2\sqrt{6}\Delta) \). If the amplitudes of the \( \sigma_{\pm} \) fields are the same, i.e., \( |g_{\mp}| = |g_{\pm}| \), and if the magnetic field is applied so that the condition \( q = -|g_{\mp}|^2/(12\Delta) \) is met, the coefficients of the first terms on the right-hand side of Eq. (43) become equal, i.e., \( 4q - |g_{\mp}|^2/(6\Delta) = -(|g_{\pm}|^2 + |g_{\pm}|^2)/(4\Delta) = 6q \). The solutions are thus obtained as

\[ \zeta_{2} = e^{-i(2p + 6q)t/\hbar} \frac{\Omega}{\sqrt{2|\Omega|^2}} \sin \sqrt{2|\Omega|^2}t, \tag{44a} \]
\[ \zeta_{0} = e^{-i6qt/\hbar} \cos \sqrt{2|\Omega|^2}t, \tag{44b} \]
\[ \zeta_{-2} = -e^{-i(-2p + 6q)t/\hbar} \frac{\Omega^*}{\sqrt{2|\Omega|^2}} \sin \sqrt{2|\Omega|^2}t. \tag{44c} \]

We note that the phase difference is always \( \chi = \chi_{2} + \chi_{-2} - 2\chi_{0} = \pi \), which is independent of the phase of the circularly polarized light and the applied magnetic field. This method is thus suitable for preparing the initial state (88).

We can check whether the prepared state has the correct relative phase \( \chi = \pi \). Rotating the spin state (25) by \( \pi/2 \) around the axis on the \( x\)-\( y \) plane as \( \zeta = e^{-i\pi S_y^2/2} \zeta \) with \( S_y = S_x \cos \phi + S_y \sin \phi \), we find that the population of each component becomes

\[ |\zeta_{2}|^2 = |\zeta_{-2}|^2 = \frac{1}{8} \left( 3 \cos^2 \theta - \sqrt{3} \sin 2\theta \cos \frac{\chi}{2} \cos \chi' \right), \tag{45a} \]
\[ |\zeta_{1}|^2 = |\zeta_{-1}|^2 = \frac{1}{2} \sin^2 \theta \sin^2 \chi', \tag{45b} \]
\[ |\zeta_{0}|^2 = \frac{1}{4} \left( \cos^2 \theta + \sqrt{3} \sin 2\theta \cos \frac{\chi}{2} \cos \chi' \right), \tag{45c} \]

where \( \chi' = 2\phi + (\chi_{2} - \chi_{-2})/2 \). Let us consider the quantity

\[ f_2^2 = 2(|\zeta_{1}|^2 + |\zeta_{1}|^2) = 1 + 2 \cos^2 \theta - \sqrt{3} \sin 2\theta \cos \frac{\chi}{2} \cos \chi', \tag{46} \]

which can be measured by the spin-dependent non-destructive imaging reported in Ref. [21]. We note that if \( \chi = \pi \), Eq. (10) is independent of \( \chi' \) and is constant, while if \( \chi \neq \pi \), Eq. (10) oscillates, since the phase \( \chi_{2} - \chi_{-2} \) rotates in the magnetic field. We can therefore confirm that the desired initial state \( \zeta_{2} \) is obtained if the result of the polarization-dependent phase-contrast imaging [21] is constant in time.

### VI. CONCLUSIONS

We have proposed a method to experimentally determine the sign of the singlet-pair energy of a spin-2 BEC from spin exchange dynamics, which determines whether the \( f = 2 \) \( ^{87}\text{Rb} \) BEC at zero magnetic field is antiferromagnetic or cyclic. We have obtained analytic solutions of the multicomponent GP equations and found that if we prepare the initial state in the magnetic sublevels \( m = 0 \), \( \pm 2 \) with the appropriate relative phase relationship, we can determine the sign of \( c_2 \) from the sign of the initial change in the spin populations. For example, in the case of the initial state in Eq. (25), we can conclude that \( c_2 \) is positive or negative if the \( m = 0 \) component first decreases or increases. We can also determine the magnitude of \( c_2 \) from the amplitude of the oscillation in the spin populations.

Since only the initial evolution for a period of \( \sim 10 \) ms is needed, we can use this method in the presence of atom loss by inelastic collisions and even in the presence of dynamical instabilities. We have numerically confirmed that this method is applicable for a trapped system with a realistic two-body loss coefficient \( K_2 \). The required condition for the magnetic field is \( |q| > \zeta_2/10 \), so our method works even when the quadratic Zeeman energy exceeds the singlet-pair energy. The initial spin state in which the populations and relative phases are controlled as Eq. (26) can be prepared by, e.g., the Raman technique as shown in Sec. VI.

We have shown that the coherent spin dynamics serve as an efficient probe for investigating spinor properties.
especially when the atomic species have a short lifetime and therefore the equilibrium state cannot be reached. The coherent spin dynamics may also reveal other equilibrium spinor properties as well as nonequilibrium spinor properties that cannot be obtained from the static equilibrium.

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APPENDIX A: DERIVATION OF THE SOLUTION

In this appendix, we solve Eq. (29) for the initial condition (25) with \( \chi_0 = \chi_2 = 0 \) and \( 0 \leq \theta \leq \pi/2 \), and derive the solution (29). Defining a new variable

\[
\zeta_0(t) \equiv e^{i(\hat{c}_z/5+4q)t/\hbar} \zeta_0(t),
\]

we can rewrite Eq. (29) as

\[
\hat{\zeta}_0 = P \zeta_0 + Q|\zeta_0|^2 \zeta_0,
\]

where

\[
P = \frac{1}{\hbar} \left( \frac{2\hat{c}_z}{5} \hat{s} - \frac{\hat{c}_z^2}{25} - 16q^2 - \frac{16\hat{c}_q}{5} \right),
\]

\[
Q = \frac{16\hat{c}_q}{5\hbar^2},
\]

with the spin-dependent energy

\[
\varepsilon_s = \frac{\hat{c}_z}{10} (2\varepsilon_s - 2c_0^2 + 4q(1 - |\zeta_0|^2)).
\]

The initial conditions for \( \zeta_0 \) are \( \zeta_0(0) = \zeta_0(0) = \cos \theta_0 \) and

\[
\dot{\zeta}_0(0) = \frac{i}{\hbar} \left( \frac{\hat{c}_z}{5} + 4q \right) \zeta_0(0)
\]

\[
- \frac{i\hat{c}_q}{\hbar} \zeta_0(0) \left[ 2e^{i\chi_2} \varepsilon_s(0)^2 + c_0^2(0) \right],
\]

where we have used Eq. (20).

We write the complex variable as \( \zeta_0(t) \equiv R(t)e^{i\phi(t)} \) with real functions \( R(t) \) and \( \phi(t) \), where \( \phi(0) = 0 \) and \( \zeta_0(0) = R(0) \). Substituting this into Eq. (A2) and taking the real and imaginary parts, we obtain

\[
\dot{R} - PR - Q R^3 - R \dot{\phi} = 0,
\]

\[
2\dot{R} \dot{\phi} + \dot{R} \phi = 0.
\]

These equations can be integrated to give

\[
\dot{R}^2 = \frac{P R^2 + Q}{2} R^4 + C_R - \frac{C_\phi}{R^2},
\]

\[
\dot{\phi} = \frac{C_\phi}{R^2},
\]

where \( C_R \) and \( C_\phi \) are constants of integration. The time derivative of \( \zeta_0 \) at \( t = 0 \), \( \dot{\zeta}_0(0) = \dot{R}(0) + i\dot{\phi}(0)R(0) \), gives

\[
\text{Re} \dot{\zeta}_0(0) = \dot{R}(0) \text{ and } \text{Im} \dot{\zeta}_0(0) = \dot{\phi}(0)R(0).
\]

The constant \( C_\phi \) is then written as

\[
C_\phi = \dot{\phi}(0) R^2(0) - z_0(0) \text{Im} \dot{\zeta}_0(0).
\]

The constant \( C_R \) is similarly obtained as

\[
C_R = \frac{z_0(0)}{2} \dot{\zeta}_0(0)^2 - \frac{Q}{2} \dot{\zeta}_0(0)^4.
\]

We introduce a new variable \( \rho \) through

\[
\dot{R}^2 = A_0 \rho^2 + B_0,
\]

where \( A_0 \) and \( B_0 \) are constants. Substituting Eq. (A13) into Eq. (A9), we obtain

\[
A_0^2 \rho^2 = \frac{Q}{2} A_0^3 \rho^4 + \left( \frac{3Q}{2} B_0 + P \right) A_0^2 \rho^2
\]

\[
+ \left( \frac{3Q}{2} B_0^2 + 2PB_0 + C_R \right) A_0
\]

\[
+ \left( \frac{Q}{2} B_0^3 + 3PB_0^2 + C_R B_0 - C_\phi^2 \right) \rho^{-2}.
\]

We determine \( B_0 \) so that the \( \rho^{-2} \) term in Eq. (A14) vanishes:

\[
\frac{Q}{2} B_0^3 + 3PB_0^2 + C_R B_0 - C_\phi^2 = 0.
\]

The right-hand side (rhs) of Eq. (A14) then becomes quartic with respect to \( \rho \). We determine \( A_0 \) so that the rhs of Eq. (A14) becomes proportional to \((1-\rho^2)(1-\nu \rho^2)^2\); the result is

\[
A_0 = \frac{1}{2Q} \left[ -3QB_0 - 2P \right. \\
\pm \sqrt{\left( -3QB_0 - 2P \right)(QB_0 + 2P) - 8QC_R} \\
- \left. \frac{Q}{4} B_0^2 - 2PB_0 + 3QB_0 \right]
\]

with

\[
\nu = -\frac{QA_0}{QA_0 + 2P + 3QB_0}.
\]

Equation (A14) thus becomes

\[
\dot{\rho}^2 = \frac{QA_0}{2\nu} (1-\rho^2)(1-\nu \rho^2).
\]
This equation can be integrated to give
\[
\int_{\rho(0)}^{\rho(t)} \frac{d\rho}{\sqrt{(1 - \rho^2)(1 - \nu \rho^2)}} = \sqrt{\frac{Q A_0}{2\nu}} t. \tag{A19}
\]
We rewrite this equation as
\[
\int_0^{\rho(t)} \frac{d\rho}{\sqrt{(1 - \rho^2)(1 - \nu \rho^2)}} = \alpha t + \beta_0, \tag{A20}
\]
where
\[
\alpha = \sqrt{\frac{Q A_0}{2\nu}}, \tag{A21}
\]
\[
\beta_0 = \int_0^{\rho(0)} \frac{d\rho}{\sqrt{(1 - \rho^2)(1 - \nu \rho^2)}}. \tag{A22}
\]
The left-hand side of Eq. (A20) is an elliptic integral of the first kind and hence \( \rho(t) = \text{sn}(\alpha t + \beta_0 | \nu) \). Thus,
\[
|\zeta_0(t)| = R(t) = \sqrt{A_0 \text{sn}^2(\alpha t + \beta_0 | \nu) + B_0}. \tag{A23}
\]
From Eqs. (A11) and (A10), the phase \( \varphi_0 \) of \( \zeta_0 \) is given by
\[
\varphi_0(t) = -\frac{1}{\hbar} \left( \varphi_2 \frac{5}{\tilde{c}_2} + 4q \right) t + \int_0^t d\tau A_0 \text{sn}^2(\alpha t + \beta_0 | \nu) + B_0 \tag{A24}\]
Similarly, we can obtain the solution for \( \zeta_2(t) \). From the form of Eq. (26) and the initial condition \( \zeta_2(0) = e^{i\chi_2} \zeta_2(0) \), we find that the relation \( \zeta_2(t) = e^{i\chi_2} \zeta_2(t) \) always holds. Hence, we consider only \( \zeta_2(t) \). Eliminating \( \zeta_0(t) \) from Eq. (26), we have the equation of motion for \( \zeta_2(t) \) as
\[
h^2 \ddot{z}_2 = \left[ \frac{2\tilde{c}_2}{5} (\epsilon s + 4q) - 16q^2 - \frac{32\tilde{c}_2 q}{5} |\zeta_2|^2 \right] \zeta_2 - i \frac{2\tilde{c}_2}{5} \hbar \zeta_2. \tag{A25}\]
The new variable \( z_2(t) = e^{i\chi_2 t/(5\hbar)} \zeta_2(t) \) reduces Eq. (A26) to
\[
\dot{z}_2 = P' z_2 + Q' |z_2|^2 z_2, \tag{A26}\]
where \( P' = \left( 2\tilde{c}_2 \epsilon s/5 - \frac{\tilde{c}_2}{25} - 16q^2 + \frac{8\tilde{c}_2 q}{5} \right)/\hbar^2 \) and \( Q' = -32\tilde{c}_2 q/(5\hbar^2) \). The initial conditions for \( z_2 \) are \( z_2(0) = \zeta_2(0) = \sin \theta_0 / \sqrt{2} \) and
\[
\dot{z}_2(0) = \frac{i\tilde{c}_2}{5\hbar} \zeta_2(0) \left[ 1 - 2\zeta_2^2(0) - e^{-i\chi_2} \zeta_2^2(0) \right] - \frac{4iq}{\hbar} \zeta_2(0). \tag{A27}\]
Equation (A26) has the same form as Eq. (A2), in which \( P \) and \( Q \) are replaced by \( P' \) and \( Q' \), and therefore the derivation from Eq. (A24) to Eq. (A27) can similarly be applied.

We consider the case of \( \chi_2 = \pi \) in the initial condition given in Eq. (25). In this case, Eq. (A10) is pure imaginary, and Eq. (A15) can be solved to give \( B_0 = z_2^2(0) = \zeta_2^2(0) \). Equation (A16) then reduces to
\[
A_0 = \left\{ \begin{array}{l}
1 - 2\zeta_2^2(0) + \frac{10q}{\tilde{c}_2}, \\
\frac{\tilde{c}_2}{10q} \zeta_2^2(0) \left[ 1 - \zeta_2^2(0) \right],
\end{array} \right.
\tag{A28}\]
where the upper and lower expressions correspond to the plus and minus signs of \( \pm \) in Eq. (A16). Using this \( A_0, \nu, \) and \( \alpha \) become
\[
\nu = \left\{ \begin{array}{l}
10q \left\{ \frac{\tilde{c}_2}{2} \left[ 1 - 2\zeta_2^2(0) \right] + 10q \right\}, \\
\frac{\tilde{c}_2}{10q} \zeta_2^2(0) \left( 1 - \zeta_2^2(0) \right),
\end{array} \right. \tag{A29}\]
\[
\alpha = \left\{ \begin{array}{l}
\frac{2\tilde{c}_2}{5\hbar} \sqrt{\zeta_2^2(0) \left[ 1 - \zeta_2^2(0) \right]}, \\
\frac{8\tilde{c}_2}{5\hbar} \left( \frac{\tilde{c}_2}{2} \left[ 1 - 2\zeta_2^2(0) \right] + 10q \right),
\end{array} \right. \tag{A30}\]
It follows from Eq. (A13) and \( B_0 = z_2^2(0) \) that \( \rho(0) = 0 \) and hence \( \beta_0 = 0 \). Using the relation
\[
\text{sn}(\alpha t | \nu) = \frac{1}{\sqrt{\nu}} \text{sn}(\sqrt{\nu} \alpha t | \nu^{-1}), \tag{A31}\]
we can show that the two expressions in Eqs. (A28), (A29), and (A30) are equivalent to each other. We used the lower expressions in Sec. III. The \( m = \pm 2 \) components \( \zeta_2(t) = -\zeta_2(t) \) have the same form as Eq. (A28), in which \( A_0, \beta_0, \) and \( B_0 \) are replaced by \( A_2, \beta_2, \) and \( B_0 \). These constants are given by
\[
A_2 = \left\{ \begin{array}{l}
\frac{1}{2} - 2 \zeta_2^2(0) - \frac{5q}{\tilde{c}_2}, \\
\frac{\tilde{c}_2}{10q} \zeta_2^2(0) \left[ 1 - 2\zeta_2^2(0) \right],
\end{array} \right. \tag{A32}\]
\[
\beta_2 = 0, \text{ and } B_2 = \zeta_2^2(0).\]
Finally, we consider the case of \( \chi_2 = 0 \) in the initial condition (25), i.e., all the components are real and positive.
We can solve Eq. (A15) also in this case to give \( B_0 = z_0^2(0) \) and hence \( \beta_0 = 0 \). The constants \( A_0, \nu, \) and \( \alpha \) reduce to

\[
A_0 = \frac{1}{2\epsilon_2 q} \left\{ q \{ \epsilon_2 [1 - 2z_0^2(0)] + 10q \} - |q| \sqrt{(\epsilon_2 + 10q)^2 - 40\epsilon_2 qz_0^2(0)} \right\},
\]

(A33)

\[
\nu = \frac{q \{ \epsilon_2 [1 - 2z_0^2(0)] + 10q \} - |q| \sqrt{(\epsilon_2 + 10q)^2 - 40\epsilon_2 qz_0^2(0)}}{q \{ \epsilon_2 [1 - 2z_0^2(0)] + 10q \} + |q| \sqrt{(\epsilon_2 + 10q)^2 - 40\epsilon_2 qz_0^2(0)}}
\]

(A34)

\[
\alpha = \sqrt{\frac{8\epsilon_2 q A_0}{\hbar^2 \nu}}
\]

(A35)

where we take the lower sign in Eq. (A16). Since \( \{ \epsilon_2 [1 - 2z_0^2(0)] + 10q \}^2 < (\epsilon_2 + 10q)^2 - 40\epsilon_2 qz_0^2(0) \), we find \( \epsilon_2 A > 0 \) and \( \nu < 0 \).

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