Formalism of rotating-wave approximation in high-spin system with quadrupole interaction

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(Received 2 June 2022; revised manuscript received 9 September 2022; accepted manuscript online 17 November 2022)

We investigate the rotating wave approximation applied in the high-spin quantum system driven by a linearly polarized alternating magnetic field in the presence of quadrupole interactions. The conventional way to apply the rotating wave approximation in a driven high-spin system is to assume the dynamics being restricted in the reduced Hilbert space. However, when the driving strength is relatively strong or the driving is off resonant, the leakage from the target resonance subspace cannot be neglected for a multi-level quantum system. We propose the correct formalism to apply the rotating wave approximation in the full Hilbert space by taking this leakage into account. By estimating the operator fidelity of the time propagator, our formalism applied in the full Hilbert space unambiguously manifests great advantages over the conventional method applied in the reduced Hilbert space.

Keywords: rotating wave approximation, quadrupole interaction, high-spin system

PACS: 03.65.–w, 76.60.Gv **DOI:** 10.1088/1674-1056/aca392

1. Introduction

Rotating wave approximation (RWA) plays an important role in many fields of quantum physics, such as magnetic resonance,[1–3] quantum optics,[4–7] condensed matter physics,[8] atomic physics,[9] etc. Specifically, Rabi model,[5,10] a model of numerous practical importance, which describes a two-level system driven by a linearly polarized ac field, needs the rotating wave approximation to get the analytic treatment. The applicability of RWA in the driven two-level system needs the conditions that the driving is near resonance and the driving strength is relatively weak,[5,7,11] otherwise the counter-rotating terms may lead to significant effects, such as the Bloch–Siegert shifts,[11] the coherent destruction of tunneling,[12,13] and so on. During recent years, many works have dedicated to devise modified RWA formalism in the driven two-level system[14–18] by taking into account effects from the counter-rotating terms.

Recently, high-spin quantum systems (with spin $I > 1/2$) attract much attention in quantum computing[19] and quantum sensing.[20,21] Since high-spin quantum systems possess much more quantum degrees of freedom, they can be harnessed to realize many advanced techniques in these quantum technologies, like sensing assisted by auxiliary qubits,[22] quantum error correction,[19] etc. Particularly, the knowledge on the dynamics of such a driven multi-level quantum system, especially on an analytic treatment, is crucial for many practical applications in quantum sensing or quantum computing, such as high-fidelity quantum state preparation,[23] high-quality quantum operations,[19,24] high-precision quantum measurements,[20] etc. However, since the quadrupole interaction always presents in a high-spin quantum system,[1,2,25] it results in difficulty to directly employ the standard RWA formalism when the high-spin system is driven by a linearly polarized ac field. In particular, strong driving is required to build fast quantum gates in quantum computation,[26] which unavoidably leads to leakage from the target transition subspace in a multi-level system. This leakage should be taken into account to correctly apply the rotating wave approximation. In this paper, we carefully investigate the rotating wave approximation in a high-spin quantum system in the presence of quadrupole interactions and propose the correct RWA formalism in such a driven multi-level quantum system.

For a high-spin system, typically a nucleus with high spin, driven by a time-dependent magnetic field in the presence of quadrupole interactions, the Hamiltonian can be written as[1,2,27]

$$H_{\rm Q}(t) = \gamma_{\rm n}\hbar \boldsymbol{B}(t)\cdot\boldsymbol{I} + \frac{e^2qQ_0}{4I(2I-1)} \times \left[3I_z^2 - I(I+1) + \frac{\eta}{2}(I_+^2 + I_-^2)\right]. \quad (1)$$

The first term corresponds to the Zeeman interaction of the nuclear spin $\boldsymbol{I}$ with the applied magnetic field $\boldsymbol{B}(t)$, where $\gamma_{\rm n}$ is the gyromagnetic ratio of the spin and $\hbar$ is the Planck constant. The second term corresponds to the quadrupole interaction for spin $I > 1/2$, where e is the elementary electric charge, Q_0 is the quadrupole moment of the nucleus, q is the

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crystal lattice field gradient, and η is the asymmetry parameter. $I_{\alpha=x,y,z}$ are the spin operators, with $I_\pm = I_x \pm \mathrm{i}I_y$. For the high-spin system driven by a linearly polarized ac magnetic field, $\boldsymbol{B}(t) = (B_1\cos(\omega t), 0, B_0)$, where B_0 is the strength of the static magnetic field, while B_1 and ω are the amplitude and frequency of the ac field, respectively. For simplicity, throughout this paper we set $\hbar = 1$ and assume the gyromagnetic ratio of the spin $\gamma_n = 1$. Also, we assume the asymmetry parameter $\eta = 0$ and the above Hamiltonian can be simplified as

$$H(t) = QI_z^2 + B_0 I_z + B_1\cos(\omega t)I_x, \tag{2}$$

where we have omitted the constant terms and denoted the quadrupole coupling strength $Q \equiv \frac{3e^2qQ_0}{4I(2I-1)}$. Typically, such a Hamiltonian describes a nuclear spin with quadrupole moment driven by external magnetic fields. However, the quadruple term can also origin from many-body interactions.[28–32] For instance, the quadrupole term in the Hamiltonian of the nitrogen-vacancy center in diamond origins from the electronic interactions.[33]

Usually, the interaction of the quantum spin system with an alternating magnetic field can be approximated by the interaction of the quantum spin system with a rotating magnetic field. This is done by decomposing the interaction of the spin with the alternating magnetic field into the co-rotating terms and the counter-rotating terms, then the counter-rotating terms are neglected due to their rapid oscillations in the interaction picture. This is the well-known standard rotating wave approximation.[1,2] For a brief review of the standard RWA applied in the high-spin system, we consider the situation that the Zeeman splitting is significantly larger than the quadrupole coupling, namely, $B_0 \gg Q$ in $H(t)$. As shown in Fig. 1(a), the effect of the quadrupole coupling is to shift the Zeeman energy level, leading to deviations of the transition frequency between nearest Zeeman levels. In fact, the standard rotating wave approximation can be directly applied for this situation. Firstly, we need to transform the original Hamiltonian [Eq. (2)] into the interaction picture (set by $H_0 = B_0 I_z + QI_z^2$),

$$\begin{aligned} H_{\mathrm{I}}(t) &= \mathrm{e}^{\mathrm{i}H_0 t}H(t)\mathrm{e}^{-\mathrm{i}H_0 t} - H_0 \\ &= \frac{B_1}{2}\mathrm{e}^{\mathrm{i}(B_0 I_z + QI_z^2 - \omega I_z)t}I_x\mathrm{e}^{-\mathrm{i}(B_0 I_z + QI_z^2 - \omega I_z)t} \\ &\quad + \frac{B_1}{2}\mathrm{e}^{\mathrm{i}(B_0 I_z + QI_z^2 + \omega I_z)t}I_x\mathrm{e}^{-\mathrm{i}(B_0 I_z + QI_z^2 + \omega I_z)t}. \end{aligned} \tag{3}$$

When the driving is near resonance $\omega \sim B_0$, and since $B_0 \gg Q$, we only keep the first (slowly oscillating) term for the RWA. We then transform back to the Schrödinger picture and the Hamiltonian after applying the standard rotating wave approximation now becomes

$$H(t) \approx QI_z^2 + B_0 I_z + \frac{B_1}{2}\mathrm{e}^{-\mathrm{i}\omega I_z t}I_x\mathrm{e}^{\mathrm{i}\omega I_z t}. \tag{4}$$

The dynamics of this Hamiltonian can be exactly solved in the rotating reference frame[34] by applying the unitary transformation $U(t) = \mathrm{e}^{-\mathrm{i}\omega I_z t}$. Accordingly, the effective Hamiltonian in this rotating reference frame becomes time-independent,

$$\begin{aligned} H_{\mathrm{eff}} &= U^\dagger(t)H(t)U(t) - \mathrm{i}U^\dagger(t)\frac{\partial}{\partial t}U(t) \\ &= (B_0 - \omega)I_z + QI_z^2 + \frac{B_1}{2}I_x. \end{aligned} \tag{5}$$

Finally, after applying the standard RWA, the time evolution operator corresponding to $H(t)$ in Eq. (2) can be written explicitly as

$$\mathcal{U}(t) \approx \mathrm{e}^{-\mathrm{i}\omega I_z t}\,\mathrm{e}^{-\mathrm{i}H_{\mathrm{eff}}t}. \tag{6}$$

However, when the quadrupole interaction dominates in $H(t)$, namely, $Q \gg B_0$, the situation changes dramatically. As shown in Fig. 1(b), the quadrupole interaction results in degeneracy of energy levels and this degeneracy is lifted by the Zeeman term. Since now transitions do not occur between nearest energy levels, the above procedure to directly apply the standard rotating wave approximation cannot be employed anymore. Alternatively, we can understand this fact from another perspective. The unitary operator, $\exp(-\mathrm{i}QI_z^2 t) = \exp[-\mathrm{i}(QI_z t)I_z]$, can be regarded as the rotation around the z-axis, but the rotation frequency and direction ($+z$-axis or $-z$-axis) are now dependent on I_z at the same time. It is this characteristic that results in the difficulty to directly employ the standard RWA procedure. In this paper, we will devote ourselves to find out the correct formalism to apply the rotating wave approximation in the driven high-spin system when the quadrupole interaction dominates.

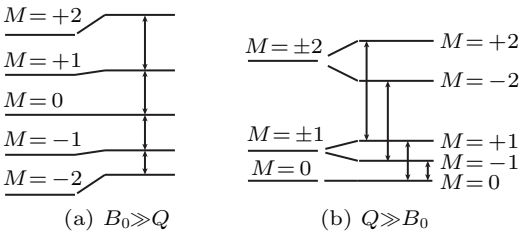


Fig. 1. Energy levels correspond to $H_0 = QI_z^2 + B_0 S_z$ for spin $I = 2$. (a) When Zeeman energy dominates, the quadrupole interaction leads to shifts of the energy levels. (b) When quadrupole interaction dominates, the Zeeman term leads to the splitting of the degeneracy. In both figures, the arrows indicate possible transitions induced by $H_1 = B_1\cos(\omega t)I_x$.

In order to provide an elaborate analysis on the rotating wave approximation for the high-spin system in the presence of quadrupole interactions, we organize our paper as follows. In Section 2, we propose our formalism to apply the rotating wave approximation in the high-spin system when the quadruploe interaction dominates. In Section 3, we discuss the conventional method to apply RWA in a multi-level quantum system. In Section 4, we compare the performance of our RWA formalism with the standard RWA procedure by calculating

the operator fidelity. Finally, we discuss problems that need to be investigated in the future and possible applications of our RWA formalism in Section 5.

2. Rotating wave approximation when quadrupole interaction dominates

We now consider the procedure to apply the rotating wave approximation in high-spin systems when the quadrupole interaction in $H(t)$ dominates, namely, $Q \gg B_0$. For half-integer spin I, since there exists transition between levels $|I, M = 1/2\rangle$ and $|I, M = -1/2\rangle$, it is difficult to find an appropriate operator to apply our RWA formalism (see below). Therefore, in this section, we will first introduce the RWA formalism for the integer high-spin and then generalize this formalism for the half-integer high-spin.

2.1. Rotating wave approximation for integer high spin

The Hamiltonian in the interaction picture in Eq. (3) can be explicitly represented as

$$\begin{aligned} & H_{\rm I}(t) \\ = & \sum_{M=-I+1}^{I} B_1' \big[(\mathrm{e}^{-\mathrm{i}(\omega_M-\omega)t} + \mathrm{e}^{-\mathrm{i}(\omega_M+\omega)t}) |I, M-1\rangle\langle I, M| \\ & + (\mathrm{e}^{\mathrm{i}(\omega_M-\omega)t} + \mathrm{e}^{\mathrm{i}(\omega_M+\omega)t}) |I, M\rangle\langle I, M-1| \big], \end{aligned} \tag{7}$$

where $B_1' = \frac{B_1}{4}\sqrt{(I+M)(I-M+1)}$ and $\omega_M = (2M-1)Q + B_0$. $|I, M\rangle$'s are the eigenstates of $\boldsymbol{I}^2$ and I_z, with M being the eigenvalue of I_z and $I(I+1)$ the eigenvalue of $\boldsymbol{I}^2$. For the rotating wave approximation, we neglect these rapidly oscillating terms, namely, terms with the exponent $\propto (nQ + \omega)$, with integer $n > 0$. After neglecting these terms, the Hamiltonian in the interaction picture becomes

$$\begin{aligned} H_{\rm I}(t) \approx & \sum_{M=1}^{I} B_1' \big[\mathrm{e}^{-\mathrm{i}(\omega_M-\omega)t} |I, M-1\rangle\langle I, M| \\ & + \mathrm{e}^{\mathrm{i}(\omega_M-\omega)t} |I, M\rangle\langle I, M-1| \big] \\ & + \sum_{M=-I+1}^{0} B_1' \big[\mathrm{e}^{-\mathrm{i}(\omega_M+\omega)t} |I, M-1\rangle\langle I, M| \\ & + \mathrm{e}^{\mathrm{i}(\omega_M+\omega)t} |I, M\rangle\langle I, M-1| \big]. \end{aligned} \tag{8}$$

We can check that, after transforming $H_{\rm I}(t)$ back to the Schrödinger picture, the Hamiltonian actually has the following form:

$$H(t) \approx QI_z^2 + B_0 I_z + \frac{B_1}{2} \mathrm{e}^{-\mathrm{i}\omega t I_z I_a} I_x \mathrm{e}^{\mathrm{i}\omega t I_z I_a}, \tag{9}$$

where we have introduced a specific operator

$$I_{\rm a} = \sum_{M=1}^{I} |I, M\rangle\langle I, M| - \sum_{M'=-I}^{-1} |I, M'\rangle\langle I, M'|. \tag{10}$$

This operator $I_{\rm a}$ is the key to obtain the correct RWA formalism for driven high-spin systems in the presence of quadrupole interactions.

Accordingly, now we can transform $H(t)$ to the rotating reference frame by applying the unitary transformation

$$U(t) = \mathrm{e}^{-\mathrm{i}\omega t I_z I_{\rm a}}. \tag{11}$$

Hence, the effective Hamiltonian in this rotating reference frame becomes time-independent,

$$\begin{aligned} H_{\rm eff} &= U^\dagger(t) H(t) U(t) - \mathrm{i} U^\dagger(t) \frac{\partial}{\partial t} U(t) \\ &= QI_z^2 - \omega I_z I_{\rm a} + B_0 I_z + \frac{B_1}{2} I_x, \end{aligned} \tag{12}$$

where we have used the fact that $[I_{\rm a}, I_z] = 0$. Finally, the time evolution operator after applying the rotating wave approximation can be written as

$$\mathcal{U}(t) \approx \mathrm{e}^{-\mathrm{i}\omega t I_z I_{\rm a}} \mathrm{e}^{-\mathrm{i} H_{\rm eff} t}. \tag{13}$$

2.2. Rotating wave approximation for half-integer high spin

Next, we consider the formalism of the rotating wave approximation when the quadrupole interaction dominates for a half-integer high-spin system. The spin angular momentum $\boldsymbol{J}$ with half-integer quantum number J, can be regarded as the addition of two angular momenta,

$$\boldsymbol{J} = \boldsymbol{I} + \boldsymbol{S}, \tag{14}$$

where $\boldsymbol{S}$ is the operator of spin $S = 1/2$ and $\boldsymbol{I}$ is the operator of integer spin I. Then, the Hamiltonian in Eq. (2) for half-integer high spin J can be rewritten as follows:

$$\begin{aligned} H(t) &= QJ_z^2 + B_0 J_z + B_1 \cos(\omega t) J_x \\ &= QI_z^2 + B_0 I_z + B_1 \cos(\omega t) I_x + QS_z^2 \\ &\quad + B_0 S_z + B_1 \cos(\omega t) S_x + 2QI_z S_z. \end{aligned} \tag{15}$$

Since QS_z^2 is a constant term, we can neglect it from the Hamiltonian. Following the procedure introduced above to apply the rotating wave approximation for integer spin I, we get the effective Hamiltonian in the rotating frame [after applying transformation $U_1(t) = \exp(-\mathrm{i}\omega t I_z I_{\rm a})$] as follows:

$$\begin{aligned} H_{\rm eff} &= QI_z^2 - \omega I_z I_{\rm a} + B_0 I_z + \frac{B_1}{2} I_x \\ &\quad + B_0 S_z + B_1 \cos(\omega t) S_x + 2QI_z S_z. \end{aligned} \tag{16}$$

Now, since the time dependence only exists in the term with spin $S = 1/2$, we can apply the standard RWA procedure. The effective Hamiltonian after applying the RWA in the rotating frame [by unitary transformation $U_2(t) = \exp(-\mathrm{i}\omega t S_z)$] becomes time-independent,

$$\begin{aligned} H_{\rm eff}' &= QI_z^2 - \omega I_z I_{\rm a} + B_0 I_z + \frac{B_1}{2} I_x \\ &\quad + (B_0 - \omega) S_z + \frac{B_1}{2} S_x + 2QI_z S_z. \end{aligned} \tag{17}$$

Eventually, the time evolution operator for the driven half-integer high-spin system can be written as

$$\mathcal{U}(t) \approx \exp(-\mathrm{i}\omega t I_z I_\mathrm{a}) \exp(-\mathrm{i}\omega t S_z) \exp(-\mathrm{i} H'_\mathrm{eff} t). \quad (18)$$

Actually, the angular momentum addition in Eq. (14) leads to $J = I + 1/2$ and $J = I - 1/2$, but we only need to calculate the dynamics in the $J = I + 1/2$ subspace. Therefore, to represent Eq. (18) in the matrix form, we have to expand it in the $J = I + 1/2$ subspace and the following relation between basis states[35] should be used:

$$\begin{aligned} &|J = I + \frac{1}{2}, M_J\rangle \\ &= \sqrt{\frac{J+M_J}{2J}} \left| S = \frac{1}{2}, M_S = \frac{1}{2} \right\rangle \otimes \left| I, M_I = M_J - \frac{1}{2} \right\rangle \\ &\quad + \sqrt{\frac{J-M_J}{2J}} \left| S = \frac{1}{2}, M_S = -\frac{1}{2} \right\rangle \otimes \left| I, M_I = M_J + \frac{1}{2} \right\rangle, \end{aligned} \quad (19)$$

where $|I, M_I\rangle$ is the eigenstate of I_z and $|S, M_S\rangle$ is the eigenstate of S_z.

3. Rotating wave approximation in the reduced Hilbert space

In this section, we discuss the conventional way[1,36] to apply the standard RWA in the high-spin system when the quadrupole interaction dominates. The resonance frequency between levels $|I, M\rangle$ and $|I, M-1\rangle$ is

$$\omega_M = (2M-1)Q + B_0, \quad (20)$$

which coincides with the resonance frequency between levels $|I, -M+1\rangle$ and $|I, -M\rangle$ when the quadrupole interaction dominates ($Q \gg B_0$). As a result, when the applied ac magnetic field is near resonance $\omega \sim |\omega_M|$ [the detuning is far from the nearest level splitting $\omega - |(2M-1)Q| \ll |(2M-3)Q|$] and the driving strength is relatively weak ($B_1 \ll \omega$), significant transitions only occur between levels $|I, M\rangle$ and $|I, M-1\rangle$, along with between levels $|I, -M+1\rangle$ and $|I, -M\rangle$. Generally, when the frequency of the applied field $\omega \sim |\omega_M|$ with $|M| > 1$, the reduced Hilbert space can be further treated as two decoupled two-level systems, since the dynamics in the $|I, M\rangle$, $|I, M-1\rangle$ subspace is decoupled from the dynamics in the $|I, -M+1\rangle$, $|I, -M\rangle$ subspace.

Besides, we should pay more attention to two exceptional cases. First, for integer spin I, when the frequency of the applied field $\omega \sim |\omega_M|$ with $|M| = 1$, the reduced Hilbert space is spanned by $|I, M = 1\rangle$, $|I, M = 0\rangle$ and $|I, M = -1\rangle$, which is an effective three-level system. Second, for half-integer spin I, when the frequency of the applied field $\omega \sim |\omega_M|$ with $|M| = 1/2$, the reduced Hilbert space is only one two-level system spanned by $|I, M = \frac{1}{2}\rangle$ and $|I, M = -\frac{1}{2}\rangle$.

3.1. Dynamics in the reduced two-level systems

Firstly, we consider the situation that the frequency of the applied ac magnetic field $\omega \sim |(2M-1)Q|$ with $|M| > 1$. As discussed above, for this situation, significant transitions take place in two decoupled two-level systems, thus the original Hamiltonian can be approximately separated into three subspace,

$$H(t) \approx H_1(t) + H_2(t) + H_n(t), \quad (21)$$

where $H_1(t)$ corresponds to the Hamiltonian projected to the $|I, M\rangle$, $|I, M-1\rangle$ subspace; $H_2(t)$ corresponds to the Hamiltonian projected to the $|I, -M+1\rangle$, $|I, -M\rangle$ subspace; $H_n(t)$ corresponds to the Hamiltonian projected to the rest of the Hilbert space. It is obvious that $[H_1(t), H_2(t)] = 0$, $[H_1(t), H_n(t)] = 0$ and $[H_2(t), H_n(t)] = 0$.

After expanding the original Hamiltonian in the $|I, M\rangle$, $|I, M-1\rangle$ subspace, we obtain

$$\begin{aligned} H_1(t) = &\left(QM^2 + B_0 M - \frac{\omega_0}{2} \right) \mathbb{1}_2 + \omega_0 S_z \\ &+ B_1 \sqrt{(I+M)(I-M+1)} \cos(\omega t) S_x, \end{aligned}$$

where $S_{\alpha=x,y,z}$ are spin operators for spin $S = 1/2$, corresponding to the two-level system spanned by $|I, M\rangle$ and $|I, M-1\rangle$. $\mathbb{1}_2$ is the 2×2 identity matrix, while $\omega_0 \equiv \omega_M = (2M-1)Q + B_0$ is the resonance frequency between levels $|I, M\rangle$ and $|I, M-1\rangle$. The first constant term in $H_1(t)$ can be neglected and we can simplify the Hamiltonian to the standard form of a quantized spin-1/2 in an alternating magnetic field,

$$H_1(t) = \omega_0 S_z + B'_1 \cos(\omega t) S_x, \quad (22)$$

where $B'_1 = B_1 \sqrt{(I+M)(I-M+1)}$ is the modified amplitude of the ac field. Using results in Subsection 2.2 (by taking spin $I = 0$), we obtain the time evolution operator corresponding to $H_1(t)$ as

$$\mathcal{U}_1(t) \approx \mathrm{e}^{-\mathrm{i}\omega t S_z} \mathrm{e}^{-\mathrm{i} H_\mathrm{eff} t}, \quad (23)$$

where the effective Hamiltonian

$$H_\mathrm{eff} = (\omega_0 - \omega) S_z + B'_1 S_x. \quad (24)$$

Through similar procedures, the evolution matrix $\mathcal{U}_2(t)$ corresponding to $H_2(t)$ in the $|I, -M+1\rangle$, $|I, -M\rangle$ subspace can be obtained as

$$\mathcal{U}_2(t) \approx \mathrm{e}^{\mathrm{i}\omega t S_z} \mathrm{e}^{-\mathrm{i} H_\mathrm{eff} t}, \quad (25)$$

along with the change of the resonance frequency $\omega_0 = (2M-1)Q - B_0$ in H_eff. Eventually, the overall time evolution operator in the whole Hilbert space can be represented as direct sum of these evolution matrices in their respective subspace,

$$\begin{aligned} \mathcal{U}(t) = &\left[\oplus_{M'=I}^{M+1} \mathrm{e}^{-\mathrm{i}(QM'^2 + B_0 M')t} \right] \\ &\oplus \mathcal{U}_1(t) \left[\oplus_{M'=M-1}^{-M+1} \mathrm{e}^{-\mathrm{i}(QM'^2 + B_0 M')t} \right] \end{aligned}$$

$$\oplus\mathcal{U}_2(t)\left[\oplus_{M'=-M-2}^{-I} \mathrm{e}^{-\mathrm{i}(QM'^2+B_0M')t}\right]. \quad (26)$$

It needs to be mentioned that, for half-integer spin I, when the frequency of the applied ac magnetic field $\omega \sim Q$, the dynamics is approximately restricted in only one two-level system spanned by $|I, M=\frac{1}{2}\rangle$ and $|I, M=-\frac{1}{2}\rangle$. Nevertheless, the time evolution operator for this case can still be obtained through similar procedures as introduced above.

3.2. Dynamics in the reduced three-level system

For integer spin I, when the frequency of the applied field $\omega \sim Q$, significant transitions occur among levels $|I, M=1\rangle$, $|I, M=0\rangle$ and $|I, M=-1\rangle$. Similar to previous discussions, when the detuning of the resonance is much smaller than the energy splitting to the nearest level, namely, $|\omega - Q| \ll 3Q$, and the driving strength is relatively weak, the dynamics can be approximately restricted to the subspace spanned by these three levels. Accordingly, the Hamiltonian can be approximately separated into two subspace, $H(t) = H_3(t) + H_n(t)$, where $H_3(t)$ corresponds to the Hamiltonian projected to the three-level subspace and $H_n(t)$ corresponds to the Hamiltonian projected to the rest of the Hilbert space. The reduced Hamiltonian in the three-level subspace is

$$H_3(t) = QS_z^2 + B_0S_z + \sqrt{\frac{I(I+1)}{2}}B_1\cos(\omega t)S_x, \quad (27)$$

where $S_{x,y,z}$ are operators for spin $S=1$, corresponding to the three-level system spanned by $|I, M=1\rangle$, $|I, M=0\rangle$ and $|I, M=-1\rangle$. Now, we can directly apply the RWA formalism obtained in Section 2, and it happens to be that $I_a = S_z$ for spin $S=1$. Straightforwardly, the time evolution operator for this subsystem is

$$\mathcal{U}_3(t) \approx \mathrm{e}^{-\mathrm{i}\omega S_z^2 t}\,\mathrm{e}^{-\mathrm{i}H_{\mathrm{eff}}t}, \quad (28)$$

where the time-independent effective Hamiltonian

$$H_{\mathrm{eff}} = (Q-\omega)S_z^2 + B_0S_z + \frac{B_1'}{2}S_x, \quad (29)$$

with the modified amplitude of the ac field $B_1' = \sqrt{I(I+1)/2}B_1$. Eventually, the overall time evolution operator in the whole Hilbert space can be represented as direct sum as follows:

$$\mathcal{U}(t) = \left[\oplus_{M=I}^{2} \mathrm{e}^{-\mathrm{i}(QM^2+B_0M)t}\right] \oplus\mathcal{U}_3(t)\left[\oplus_{M=-2}^{-I} \mathrm{e}^{-\mathrm{i}(QM^2+B_0M)t}\right]. \quad (30)$$

4. Comparison of the operator fidelity

In order to evaluate the performance of our RWA formalism compared to the conventional RWA in Section 3, we calculate the quantum operator fidelity, which is defined as[37]

$$F = \left|\frac{1}{N}\mathrm{Tr}\left[\mathcal{U}^{\dagger}(t)U(t)\right]\right|^2. \quad (31)$$

Here $\mathcal{U}(t)$ is the time evolution operator after applying our RWA formalism in Section 2 (where $N = 2I+1$ is the dimension of the corresponding operator matrix) or the conventional RWA in Section 3. $U(t)$ is the propagator obtained by exactly solving the time-dependent Schrödinger equation[38]

$$\mathrm{i}\frac{\partial U(t)}{\partial t} = H(t)U(t), \quad (32)$$

using the Runge–Kutta method with the initial condition $U(0) = \mathbb{1}$. Basically, we investigate effects of the detuning and the driving strength on the performance of these RWA formalisms. The result is presented in Fig. 2, where the value of the operator fidelity is averaged over ten π-rotation cycles, namely, the evolution time is $10T_\pi$. The characteristic time, $T_\pi = \pi/[\sqrt{\frac{1}{2}I(I+1)}B_1]$, is defined as the the time needed for a π-rotation when the conventional RWA is applied on resonance in the reduced Hilbert space (see Section 3).

In Fig. 2(a), the driving strength of the ac magnetic field is fixed at a relatively strong value $B_1 = 0.1Q$, while the frequency of the ac magnetic field is varied around the on-resonant frequency $\omega \sim Q$. Both RWA formalisms achieve the best performance when the driving is close to the exact resonance, which is as expected. Furthermore, it is clear that our RWA formalism applied in the full Hilbert space always shows much higher operator fidelity than the conventional RWA applied in the reduced Hilbert space. In Fig. 2(b), the frequency of the ac magnetic field is fixed at a nearly on-resonant value, $\omega = Q$, while the driving strength of the ac magnetic field is varied. The result shows that the performance of both RWA formalisms deteriorates when the driving strength increases. Again, it is obvious that our RWA formalism applied in the full Hilbert space always outperforms the conventional RWA applied in the reduced Hilbert space. These results indicate that, when the driving strength is relatively strong or the driving frequency is off-resonant, transitions outside the target resonance subspace (the reduced Hilbert space) or transitions between the target resonance subspace with the outside subspace cannot be neglected. As a result, the rotating wave approximation should better be applied in the full Hilbert space using our RWA formalism as proposed in Section 2.

As can be seen from Fig. 1, the quadrupole interaction tends to result in differences in transition frequencies. However, when the amplitude of the applied static magnetic field B_0 is increased, this transition frequency difference becomes less significant, and eventually ($B_0 \gg Q$) reaching nearly identical transition frequencies. Actually, the discrepancy in transition frequencies is beneficial for the employment of the conventional RWA in the reduced Hilbert space. This is verified in Fig. 2(c), where the frequency of the ac magnetic field is set to be on resonance with the transition, $|I=3, M=1\rangle \leftrightarrow$

$|I=3,M=0\rangle$. The result clearly shows that, the operator fidelity for the conventional RWA decreases significantly as B_0 increases, due to the fact that the difference in resonance frequencies between transitions $|I=3,M=1\rangle \leftrightarrow |I=3,M=0\rangle$ and $|I=3,M=-2\rangle \leftrightarrow |I=3,M=-1\rangle$ gets smaller. Meanwhile, as can be seen from Fig. 2(c), our RWA formalism applied in the full Hilbert space is robust to the change of the static magnetic field, since the leakage from the target resonance subspace is always taken into account.

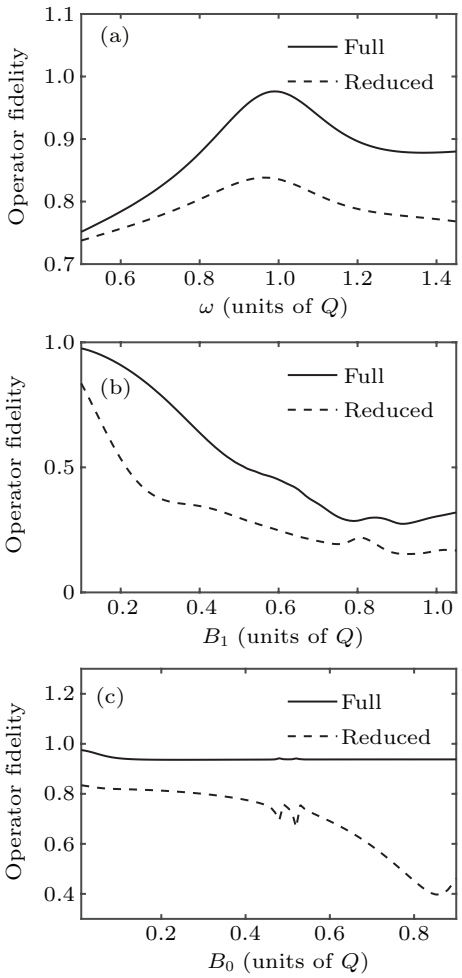


Fig. 2. Comparison of the averaged operator fidelity after applying the rotating wave approximation for spin $I=3$ using our formalism in the full Hilbert space (solid lines), or using the conventional method in the reduced Hilbert space (dashed lines). The ac magnetic field is applied to primarily induce transitions between $|I=3,M=\pm1\rangle$ and $|I=3,M=0\rangle$. (a) Operator fidelity as a function of the frequency of the ac magnetic field, with fixed amplitude of the ac magnetic field $B_1=0.1Q$ and the static magnetic field $B_0=0.02Q$. (b) Operator fidelity as a function of the strength of the ac magnetic field, with fixed driving frequency ($\omega=Q$). The static magnetic field $B_0=0.02Q$. (c) Operator fidelity as a function of the applied static magnetic field. The amplitude and frequency of the ac magnetic field are $B_1=0.1Q$ and $\omega=Q+B_0$, respectively.

5. Discussion

We now give a brief discussion on one of the possible applications of our RWA formalism in quantum sensing.[20,39,40] The dynamics of the driving high-spin system can be directly employed to estimate various parameters in the Hamiltonian, like the frequency ω or the amplitude B_1 of the driving field. We can use the explicit analytic form of the time evolution operator obtained from our RWA formalism to calculate the so-called transformed local generator,[41] $G_\lambda \equiv i\mathcal{U}^\dagger(t)\frac{\partial}{\partial\lambda}\mathcal{U}(t)$, where λ is the parameter to be estimated. Thus, we can estimate the sensitivity via the dynamic quantum Fisher information F_λ,[42] which is bounded by $F_\lambda \leq (E_{\max}-E_{\min})^2$, where $E_{\max}$ and $E_{\min}$ are the maximal and minimal eigenvalues of G_λ, respectively.

Due to its abundance on degrees of freedom, high-spin quantum system exhibits great advantages in quantum computing[19] and quantum sensing.[20,32,43] Specifically, strong driving is required to build fast quantum gates,[26] where our RWA formalism should be employed to take into account the leakage from the target resonance subspace.

In addition to the high-spin nuclear system, the Hamiltonian of a number of quantum many-body systems resembles the form of quadrupole interactions. For instance, the Lipkin–Meshkov–Glick model,[28,29] the two-mode BEC[30–32] and the spin-squeezing Hamiltonian.[44] Thus, the RWA formalism developed in this paper can be directly applied in these quantum systems. Although our RWA formalism works for the classical ac field, just like in quantum optics,[5,7] it may be generalized for quantum fields as well. Besides, our RWA formalism for the high-spin system may be further improved by taking into effects from the counter-rotating terms and this will be explored in the future.

Acknowledgements

Project supported by the National Key Research and Development Program of China (Grant Nos. 2017YFA0304202 and 2017YFA0205700), the National Natural Science Foundation of China (Grant Nos. 11875231 and 11935012), and the Fundamental Research Funds for the Central Universities (Grant No. 2018FZA3005).

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