Selective Control of the States of a Three-Level Quadrupole Nucleus by Means of Nonselective RF Pulses

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The scheme for obtaining the composite operator of the selective rotation from nonselective RF pulses separated by intervals of free evolution has been proposed for quadrupole nuclei. Using the example of three levels, it has been shown that the rotation by this operator is performed with an accuracy comparable with the accuracy of a simple selective RF pulse but in a shorter amount of time.

INTRODUCTION

In addition to two-level systems (qubits) [1], d level quantum systems (qudits) [2–9], which are more widespread in nature, are recently discussed as elements for a quantum computer. Nuclei with spin $I > 1/2$ in a high magnetic field are among these systems; the interaction between the quadrupole moments of these nuclei and the gradient of a crystal field leads to the formation of $2I + 1$ nonequidistant levels [2–5]. The state of such a nucleus can be controlled by means of pulses of the RF magnetic field. For this reason, such systems are convenient for studying quantum computations on qudits.

Selective transitions between certain levels are necessary for implementing quantum computations. For engineering reasons, experiments are performed on liquid crystals [3–5], where the gradient of the crystal field is small; hence, the difference between the frequencies of the transitions between different levels is also small. For this reason, the duration of selective RF pulses increases and becomes comparable with spin–lattice relaxation (decoherence). For systems of two spins $I = 1/2$ with close Larmor frequencies, a method was developed (see, e.g., [10]) for selective rotations by means of a sequence of short nonselective RF pulses separated by the interval of free evolution. This method is inapplicable for nonequidistant levels of the single spin that are due to the quadrupole interaction quadratic in the spin projection operators. For such levels, we propose a method based on the effective Hamiltonian method and demonstrate it in application to three nuclear levels with $I = 1$.

NUCLEAR SPIN IN THE MAGNETIC FIELD

The Hamiltonian of the nucleus with spin $I = 1$ in the high magnetic field B_0 has the form [11]

$$H_0 = -\omega_0 I_z + H_q, \quad H_q = q\left(I_z^2 - \frac{2}{3}\right), \quad (1)$$

where $\omega_0 = \gamma B_0$ is the Larmor spin precession frequency, q is the coupling constant of the quadrupole moment of the nucleus with the gradient of the crystal field, and I_z is the spin projection operator onto the direction of the constant external field (the Z axis). The energy is measured in frequency units; i.e., we set $\hbar = 1$. The quadrupole interaction H_q gives rise to the formation of three nonequidistant levels with different I_z values:

$$|I_z = 1\rangle = |0\rangle; \quad |I_z = 0\rangle = |1\rangle;$$
$$|I_z = -1\rangle = |2\rangle. \quad (2)$$

These states are taken as basis states of the qutrit.

An RF pulse is obtained when an RF field with amplitude B_1 and frequency ω is switched on for a finite time $t_r \gg 1/\omega$. The time evolution of the state in the reference frame rotating with frequency ω [11] is described by the evolution operator

$$U(t) = e^{-iHt} \quad (3)$$

with the time-independent effective Hamiltonian

$$H = H_q + (\omega - \omega_0)I_z - \frac{1}{2}\Omega(e^{-i\varphi}I^- + e^{i\varphi}I^+). \quad (4)$$

Here, $I^\pm = I_x \pm iI_y$ are the increasing and decreasing operators, respectively, and $\Omega = \gamma B_1$. The RF field phase φ determines the field direction in the rotating coordi-

nate system. The field for $\varphi = \pi$ ($\varphi = 0$) and $\varphi = \pi/2$ ($-\pi/2$) is directed along the X ($-X$) and Y ($-Y$) axes, respectively.

SELECTIVE ROTATION BY MEANS OF SELECTIVE PULSES

The state of the qutrit is changed by means of the unitary operators represented by 3×3 matrices in basis (2). Let us define the operator of the selective rotation about the axis between the qutrit states m and n as

$$R_\alpha^{m-n}(\theta) = e^{-i\theta B_\alpha^{m-n}}, \quad \alpha = x, y, \tag{5}$$

where

$$B_x^{0-1} = \frac{1}{2}\begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad B_x^{1-2} = \frac{1}{2}\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix},$$

$$B_y^{0-1} = \frac{1}{2}\begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad B_y^{1-2} = \frac{1}{2}\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{bmatrix}. \tag{6}$$

For example, the matrix of the rotation about the Y axis between levels 1 and 2 has the form

$$R_y^{1-2}(\theta) = e^{-i\theta B_y^{1-2}} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos\frac{\theta}{2} & -\sin\frac{\theta}{2} \\ 0 & \sin\frac{\theta}{2} & \cos\frac{\theta}{2} \end{bmatrix}.$$

The action of an individual RF pulse is specified by evolution operator (3) with Hamiltonian (4). In order to obtain selective rotation operator (5) between states m and n, it is necessary to choose the RF field frequency equal to the frequency of transitions between the corresponding energy levels

$$\omega^{0-1} = \omega_0 - q; \quad \omega^{1-2} = \omega_0 + q, \tag{7}$$

and pulse duration

$$t_r = \theta/\sqrt{2}\Omega. \tag{8}$$

In this case, the field amplitude must be much smaller than the difference between the resonance frequencies of various transitions ($\Omega \ll q$).

Gaussian, rather than rectangular, selective pulses are used in experiments [4]:
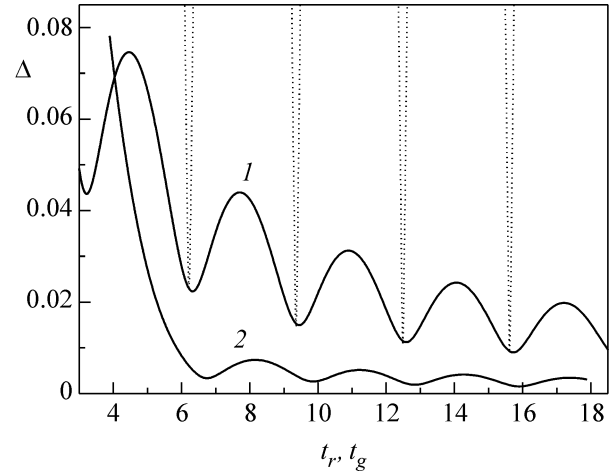
$$\Omega(t) = \Omega\exp\{-g^2(2t/t_g - 1)^2\}, \tag{9}$$

Fig. 1. Error of the selective rotation R_y^{1-2} ($\pi/2$) vs. the duration of the RF pulse (in the units of $1/q$). The solid line is the error of the absolute values Δ_2 for rectangular pulse (*1*) and Gaussian pulse (*2*) with the parameters given by Eq. (10). The dashed line is the total error of Δ_1 for the Gaussian pulse.

where t_g is the pulse duration and g is the limiting exponent. For example, for $g = 1.5$, $\exp\{-g^2\} = 0.105$ and

$$t_g = 1.752\frac{\theta}{\sqrt{2}\Omega}. \tag{10}$$

A Gaussian pulse is obtained by composing a package of narrow rectangular pulses whose amplitude varies according to Eq. (9).

Nonresonant transitions between the level with energy $2q$ and two other levels with close energies $\pm\Omega/\sqrt{2}$ [we omit the term $-2q/3$ in Eq. (3) leading to an unobservable common phase factor] occur simultaneously with resonant transitions. These transitions lead to an error. An additional contribution to the error comes from the phase factor $\exp\{-it2q\}$ of the nonresonant level, because it corresponds to one in the operator $R_\alpha^{m-n}(\theta)$. The error shown in Fig. 1 is estimated both with and without the inclusion of the phase of matrix elements from the formulas

$$\Delta_1 = \frac{1}{3}\sqrt{\sum_{i,j}|U_{ij} - U_{ij}^{\text{theor}}|^2},$$

$$\Delta_2 = \frac{1}{3}\sqrt{\sum_{i,j}||U_{ij}| - |U_{ij}^{\text{theor}}||^2}, \tag{11}$$

where U_{ij}^{theor} are the matrix elements of the ideal operator $R_\alpha^{m-n}(\theta)$ given by Eq. (5) and U_{ij} are the matrix elements of the evolution operator given by Eq. (3) that are calculated with the use of the MATLAB package.

Parameter values for obtaining B_α^{m-n} matrices

	φ	ψ	$\tau_1 q$	$\tau_2 q$	ξ	η
B_x^{0-1}	$\dfrac{\pi}{2}$	$\dfrac{\pi}{4}$	$\dfrac{1}{2\sqrt{2}}$	$\dfrac{1}{\sqrt{2}}$	$\dfrac{1}{2\sqrt{2}}$	0
B_{-x}^{0-1}	$\dfrac{\pi}{2}$	$\dfrac{\pi}{4}$	$\dfrac{1}{2\sqrt{2}}$	$\dfrac{1}{\sqrt{2}}$	$-\dfrac{1}{2\sqrt{2}}$	0
B_x^{1-2}	$\dfrac{\pi}{2}$	$-\dfrac{\pi}{4}$	$\dfrac{1}{2\sqrt{2}}$	$\dfrac{1}{\sqrt{2}}$	$\dfrac{1}{2\sqrt{2}}$	0
B_{-x}^{1-2}	$\dfrac{\pi}{2}$	$-\dfrac{\pi}{4}$	$\dfrac{1}{2\sqrt{2}}$	$\dfrac{1}{\sqrt{2}}$	$-\dfrac{1}{2\sqrt{2}}$	0
B_y^{0-1}	$-\dfrac{\pi}{4}$	$\dfrac{\pi}{2}$	$\dfrac{1}{\sqrt{2}}$	$\dfrac{1}{2\sqrt{2}}$	0	$\dfrac{1}{2\sqrt{2}}$
B_{-y}^{0-1}	$-\dfrac{\pi}{4}$	$-\dfrac{\pi}{2}$	$\dfrac{1}{\sqrt{2}}$	$\dfrac{1}{2\sqrt{2}}$	0	$-\dfrac{1}{2\sqrt{2}}$
B_y^{1-2}	$\dfrac{\pi}{4}$	$\dfrac{\pi}{2}$	$\dfrac{1}{\sqrt{2}}$	$\dfrac{1}{2\sqrt{2}}$	0	$\dfrac{1}{2\sqrt{2}}$
B_{-y}^{1-2}	$\dfrac{\pi}{4}$	$-\dfrac{\pi}{2}$	$\dfrac{1}{\sqrt{2}}$	$\dfrac{1}{2\sqrt{2}}$	0	$-\dfrac{1}{2\sqrt{2}}$

Oscillations with high frequency $\omega_q \sim 2q$ are observed against the monotonic increase in the error. The error is minimal if nonresonant levels are rotated by an angle multiple to 2π in time $t_r(t_g)$. The error of the absolute values of the matrix elements at the minima is smaller than the total error; therefore, a small phase difference between these elements holds. It is seen that the error of the Gaussian pulse is smaller than the error of the rectangular pulse, but the Gaussian pulse cannot be made shorter than $\sim 6/q$, because the error begins to abruptly increase for shorter pulses.

SELECTIVE ROTATION BY MEANS OF NONSELECTIVE PULSES

The nonselective rotation operator acts simultaneously on three levels:

$$P_\alpha(\theta) = e^{-i\theta I_\alpha}. \tag{12}$$

For example, the rotation matrix about the Y axis has the form

$$P_\alpha(\theta) \equiv e^{-i\theta I_y} = \frac{1}{2}\begin{bmatrix} 1+\cos\theta & -\sqrt{2}\sin\theta & 1-\cos\theta \\ \sqrt{2}\sin\theta & 2\cos\theta & -\sqrt{2}\sin\theta \\ 1-\cos\theta & \sqrt{2}\sin\theta & 1+\cos\theta \end{bmatrix}.$$

To implement the operator given by Eq. (12), the amplitude of the RF field in Eq. (3) must be much larger than the difference between the resonance frequencies of various transitions, i.e., $\Omega \gg q$. At $\omega = \omega_0$, for the RF pulse corresponding to Eq. (12), it follows from Eq. (3) that

$$\{\theta\}_\alpha = e^{-it_p(H_q - \Omega I_\alpha)}, \tag{13}$$

where $t_p = \theta/\Omega$ is the pulse duration.

In order to obtain the selective transition on qutrit states, we must construct an effective Hamiltonian in Eq. (3) with the form of matrices B_α^{m-n} (6). This Hamiltonian must include operators describing the allowable interactions on the system. Let us introduce the notation

$$A = e^{-i\varphi I_x}(H_q\tau_1)e^{i\varphi I_x};$$
$$B = e^{-i\psi I_y}(H_q\tau_2)e^{i\psi I_y}; \quad C = \xi I_x + \eta I_y, \tag{14}$$

where the first two operators can be obtained from the free-evolution operators and nonselective-rotation operators with the use of the property of exponential operators

$$e^{-i\varphi I_x}e^{iHt}e^{i\varphi I_x} = e^{e^{-i\varphi I_x}Hte^{i\varphi I_x}}, \tag{15}$$

and the third operator in Eqs. (14) can be obtained by means of the RF field. Equating the sum of the matrices of operators (14) to Eq. (6), we arrive at the system of nine equations

$$B_\alpha^{m-n} = H_{\mathrm{eff}}\tau$$

$$= \begin{bmatrix} \frac{1}{6}((3\cos^2\varphi - 1)q\tau_1 + 3(\cos^2\psi - 1)q\tau_2) \\[2mm] \frac{\sqrt{2}}{2}i(q\tau_1\sin\varphi\cos\varphi - \eta + i(q\tau_2\sin\psi\cos\psi + \xi)) \\[2mm] -\frac{1}{2}((1-\cos^2\varphi)q\tau_1 + (\cos^2\psi - 1)q\tau_2) \end{bmatrix}$$

$$\frac{\sqrt{2}}{2}i(-q\tau_1\sin\varphi\cos\varphi + \eta + i(q\tau_2\sin\psi\cos\psi + \xi))$$

$$-\frac{1}{3}((3\cos^2\varphi - 1)q\tau_1 + (3\cos^2\psi - 1)q\tau_2)$$

$$-\frac{\sqrt{2}}{2}i(q\tau_1\sin\varphi\cos\varphi + \eta + i(q\tau_2\sin\psi\cos\psi - \xi))$$

$$-\frac{1}{2}((1-\cos^2\varphi)q\tau_1 + (\cos^2\psi - 1)q\tau_2)$$

$$-\frac{\sqrt{2}}{2}i(-q\tau_1\sin\varphi\cos\varphi - \eta + i(q\tau_2\sin\psi\cos\psi - \xi)).$$

$$\frac{1}{6}((3\cos^2\varphi - 1)q\tau_1 + (3\cos^2\psi - 1)q\tau_2)$$

The solution of this system provides desired parameter values presented in the table. Note that the parameters τ_1 and τ_2 determining the free-evolution times must be

always positive; hence, the selective rotations at positive and negative angles are implemented with different values of the RF field parameters.

Thus, at parameters presented in the table, we obtain the selective rotation matrices in the form of the sum

$$\theta B_\alpha^{m-n} = \theta(A + B + C). \quad (16)$$

Since the operators in this expression do not commute with each other, in order to obtain the desired result, we use the Trotter–Suzuki formula [12] for the exponential operators:

$$(e^{-i\theta A/2n} e^{-i\theta B/2n} e^{-i\theta C/n} e^{-i\theta B/2n} e^{-i\theta A/2n})^n$$
$$= e^{-i\theta(A + B + C)} + O((\theta/n)^3). \quad (17)$$

In view of Eq. (16), the presented product of the operators in the limit $n \longrightarrow \infty$ converges to the ideal selective rotation given by Eq. (5).

The product of the operators repeated n times on the left-hand side of Eq. (17) can be obtained by means of RF pulses separated by intervals of free evolution. In particular, the sequence for the selective rotation about the Y axis is written in the form

$$\{\varphi\}_{-x} \xleftarrow{t_1/2} \{\varphi\}_x \{\psi\}_{-y} \xleftarrow{t_2/2} \{\psi\}_y \{\delta_y\}_y \{\psi\}_{-y}$$
$$\xleftarrow{t_2/2} \{\psi\}_y \{\varphi\}_{-x} \xleftarrow{t_1/2} \{\varphi\}_x = \{\varphi\}_{-x} \quad (18)$$
$$\xleftarrow{t_1/2} \{\varphi\}_x \{\psi\}_{-y} \xleftarrow{t_2/2} \{\delta_y\}_y \xleftarrow{t_1/2} \{\psi\}_y \{\varphi\}_{-x} \xleftarrow{t_1/2} \{\varphi\}_x.$$

Here, $t_1 = \theta\tau_1/n$, $t_2 = \theta\tau_2/n$, $\delta_x = \theta\xi/n$, $\delta_y = \theta\eta/n$, and $\xleftarrow{t} \equiv e^{-it H_q}$ means an interval of free evolution. In transformation (18), we use the property of the rotations about one (Y) axis and remove two pulses. A similar truncated sequence can be obtained for the X rotation if operator A changes to B and vise versa.

Formulas (15)–(17) are strictly valid when ideal nonselective rotation operators (12) are used. In the real experiment, these operators are obtained by means of the evolution operator given by Eq. (13). The presence of the quadrupole interaction along with the RF field leads to errors disappearing only in the limit $\Omega \longrightarrow \infty$. In particular, since the quadrupole interaction does not change sign when the rotation angle changes sign (the direction of the RF field changes), the condition

$$\{\theta\}_\alpha \{\theta\}_{-\alpha} = 1,$$

and, therefore, Eq. (15) are not satisfied. Numerical error (11) of operator (17) implemented by means of the sequence of RF pulses (18) is shown in Fig. 2. A feature of sequence (18) is the presence of RF pulses that are introduced for transformation (15) of the Hamiltonian H_q and thereby do not undergo scaling with changing n. The number of such pulses increases proportionally to n, and, correspondingly, the total error caused by H_q in Eq. (13), as well as the sequence duration, increases. This is seen in Fig. 2. Therefore, in order to reduce the error associated with the noncom-
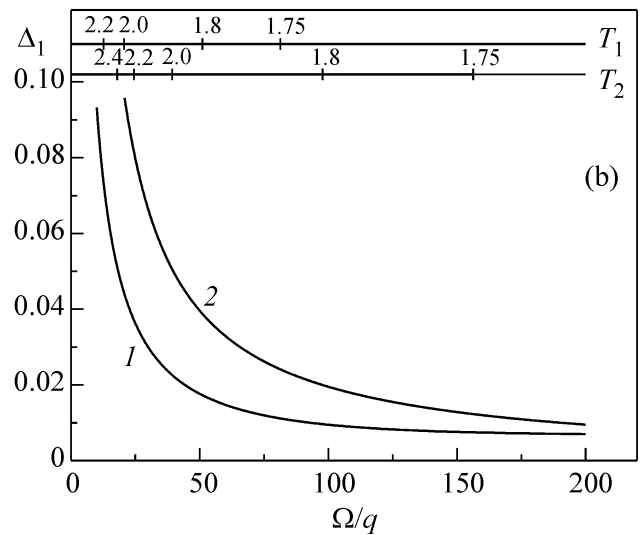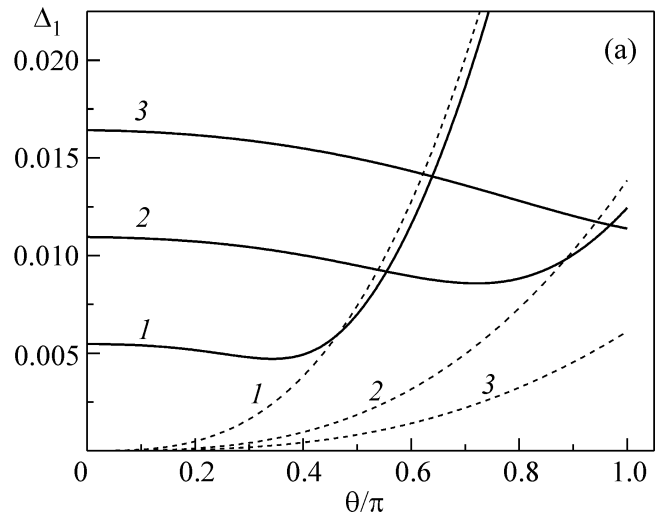
Fig. 2. Total error Δ_1 for the composite selective rotation operator $R_y^{1-2}(\theta)$ vs. (a) the angle θ at $\Omega = 200q$ and (b) the amplitude of the RF field at $\theta = \pi/2$. The repetition number n in Eq. (17) is given as the digits near the lines. The dashed lines in (a) show the same error after the change of nonselective RF pulses (13) to the ideal rotation operators given by Eq. (12). The upper scale in (b) shows the total duration T_n of the operator (in the units of $1/q$), which is equal to the sum of the durations of RF pulses and intervals of free evolution.

mutativity of the operators in Eq. (17), it is necessary to sufficiently increase the amplitude of the RF field.

The comparison of the results (shown in Figs. 1 and 2) of two methods of the implementation of selective rotations shows that, in the case of the weak quadrupole interaction, the rotation with a comparable accuracy can be implemented by means of a composite selective pulse in time $T \sim 1/q$, which is much smaller than the duration of a simple selective pulse $t_r \sim 1/\Omega \gg 1/q$. Note that, according to the calculation, the gain in time for the π pulse is larger than that for the $\pi/2$ pulse shown

above. The operator proposed in this study can be used to implement more complex quantum algorithms, e.g., the quantum Fourier transform [6, 9].

In conclusion, note that the proposed method of obtaining the composite selective pulse can be extended to quadrupole nuclei with many levels, as well as to the electron levels of atoms and ions in the crystal field.

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