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ATOMS, MOLECULES, OPTICS

Selective Control of the States of Multilevel Quantum Systems Using Nonselective Rotation Operators

V. E. Zobov[a], * and V. P. Shauro[b]

[a] *Kirensky Institute of Physics, Russian Academy of Sciences, Siberian Branch, Krasnoyarsk, 660036 Russia*
[b] *Siberian Federal University, Krasnoyarsk, 660041 Russia*
**e-mail: rsa@iph.krasn.ru*

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Abstract—We have calculated the sequences of nonselective rotation operators separated by intervals of free evolution that perform selective rotations between adjacent levels in systems with three, four, five, and six nonequidistant levels. We have numerically simulated the realization of the calculated sequences for quadrupole nuclei with corresponding spins controlled by intense nonselective radio-frequency (RF) pulses and investigated the dependences of the realization error on the parameters of external and internal interactions. To reduce the error when the RF field is not strong enough, we have found composite nonselective RF pulses consisting of five simple ones. We show that the error of the composite selective rotation operator can be reduced significantly in comparison to the error of a simple single selective pulse.

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1. INTRODUCTION

The development of general rules for controlling quantum systems is one of the problems of modern physics. The most rapid increase in interest to this problem is associated with its applications for controlling chemical reactions and building quantum computers [1, 2]. Selective operators that change the states of two chosen levels are commonly used in manipulating the states of multilevel quantum systems with a nonequidistant energy spectrum [3–11]. The most straightforward way of implementing such an operation is to tune the frequency of the external field to the resonance frequency of the transition between the chosen levels. The amplitude of this field should be much smaller than the frequency difference between the necessary resonance and unnecessary nonresonance transitions. The duration of the operation has to be increased when the amplitude decreases, but the time of the experiment is limited by the decoherence time.

One of the methods for reducing the operation time can be the organization of a selective operator using short intense nonselective pulses separated by intervals of free evolution under the internal interaction responsible for the level nonequidistance. As applied to quantum computing using NMR methods, this approach has been demonstrated for systems of two spins $I = 1/2$ with close Larmor frequencies (see, e.g., [12]). The proposed sequences are unsuitable for the nonequidistant levels of the single spin attributable to a quadrupole interaction quadratic in spin projection operators. For them, we [13] proposed a method based on the method of an effective Hamiltonian [14, 15] and demonstrated its work for three levels of a nucleus with spin $I = 1$. The demand for such schemes is related to NMR experiments on quadrupole nuclei in liquid-crystalline matrix [5–7]. The rapid spatial motion of molecules narrows individual lines in the NMR spectrum, causing a significant increase in decoherence time and creating favorable conditions for quantum computing. However, in this case, the crystal field gradient on a quadrupole nucleus also decreases, causing a decrease in level nonequidistance (the resonance frequency difference between the individual lines in the NMR spectrum) and creating difficulties in applying simple selective radiofrequency (RF) pulses.

In this paper, we extend our approach to four-, five-, and six-level systems (with spins 3/2, 2, and 5/2 in accordance with the formula $d = 2I + 1$; d is the number of levels in the system). To reduce the error from quadrupole interaction when the spin is rotated by a nonselective RF pulse, we found an equivalent sequence of five RF pulses for this rotation. In contrast to the previously known case [15, 16], our composite pulse reduces not only the amplitude error but also the phase one, which is important in quantum computing.

The paper is structured as follows. The construction of an effective Hamiltonian is described in Section 2. The method of its implementation using RF pulses and error reduction methods are investigated in Section 3.

The results of our numerical simulations are contained in Section 4. A composite nonselective pulse is constructed in the Appendix.

2. THE METHOD OF CONSTRUCTING AN EFFECTIVE HAMILTONIAN FOR THE COMPOSITE SELECTIVE ROTATION OPERATOR

The rotation of the magnetic (spin) moment through an angle θ about an α axis is specified by the operator

$$\{\theta\}_\alpha = \exp(-i\theta I_\alpha), \tag{2.1}$$

where I_α is the spin projection operator onto the α axis. In particular, the projection operators are transformed by the rotation according to the formulas [14, 17]

$$\begin{aligned} \exp(-i\theta I_x)I_z\exp(i\theta I_x) &= I_z\cos\theta - I_y\sin\theta, \\ \exp(-i\theta I_y)I_z\exp(i\theta I_y) &= I_z\cos\theta + I_x\sin\theta. \end{aligned} \tag{2.2}$$

The relations for other projections can be obtained using a cyclic change of variables. These formulas are valid for any spin.

The selective rotation operator $R_\alpha^{m-n}(\theta)$ of the two states corresponding to levels m and n through an angle θ about an α axis for a d-level quantum system is represented by a $d \times d$ matrix:

$$R_\alpha^{m-n}(\theta) = \begin{bmatrix} E_m & 0 & 0 & 0 & 0 \\ 0 & \cos\frac{\theta}{2} & 0 & -ie^{-i\varphi}\sin\frac{\theta}{2} & 0 \\ 0 & 0 & E_{n-m-1} & 0 & 0 \\ 0 & -ie^{i\varphi}\sin\frac{\theta}{2} & 0 & \cos\frac{\theta}{2} & 0 \\ 0 & 0 & 0 & 0 & E_{d-n-1} \end{bmatrix}. \tag{2.3}$$

Here, E_k is a unit matrix of dimension k. The phase φ defines the rotation axis. The rotation about the x ($\alpha = x$, x rotation) and y ($\alpha = y$, y rotation) axes corresponds to $\varphi = 0$ and $\pi/2$, respectively. Operator (2.3) can be written in exponential form,

$$R_\alpha^{m-n}(\theta) = \exp(-i\theta B_\alpha^{m-n}), \tag{2.4}$$

where the exponent contains a matrix in which only two elements, B_{ij} and B_{ji} ($i = m + 1$, $j = n + 1$), are nonzero.

To be able to make selective transformations, suppose that the energy level nonequidistance of the quantum magnetic moment in a strong constant magnetic field B_0 is realized by an interaction with a Hamiltonian

$$H_q = q\left(I_z^2 - \frac{1}{3}I(I+1)\right), \tag{2.5}$$

where q is the coupling constant and I is the nuclear spin. The interaction of the quadrupole moment of the nucleus with the crystal field gradient and the spin–orbit coupling of electrons in axially symmetric cases have such a form [17]. We will measure the energy in units of the angular frequency and set $\hbar = 1$.

To obtain the selective rotations (2.4) using the nonselective operators (2.1), we should transform the operator H_q so as to obtain an effective (average) Hamiltonian coincident with B_α^{m-n} (more precisely, $H_{\text{eff}}t = \theta B_\alpha^{m-n}$) from it. This problem can be solved by many methods. Let us generalize the method that we suggested previously for a three-level system [13] to systems with more than three levels.

For our system, the transitions only between the adjacent levels in whose rotation operators only the off-diagonal elements closest to the main diagonal $B_{i,\,i+1}$ are nonzero are permitted under single selective RF pulses. The matrices I_x and I_y and certain combinations of the matrices derived from Hamiltonian (2.5) by transformation (2.2) have such properties:

$$\begin{aligned} K_x\left(\frac{\pi}{2}\right) + 2K_y\left(\frac{\pi}{4}\right) &= I_zI_x + I_xI_z = M_x, \\ 2K_x\left(\frac{\pi}{4}\right) + K_y\left(\frac{\pi}{2}\right) &= -(I_zI_y + I_yI_z) = M_y, \end{aligned} \tag{2.6}$$

where

$$\begin{aligned} K_x(\psi) &= \{\psi\}_x H_q \{\psi\}_{-x}, \\ K_y(\psi) &= \{\psi\}_y H_q \{\psi\}_{-y}. \end{aligned} \tag{2.7}$$

As will be shown below, these operators can be realized in practice using nonselective RF pulses separated by intervals of free evolution.

Using the evolution operator of the system under interaction (2.5), we can change the phases of the matrix elements by a value dependent on the state:

$$e^{-itH_q}M_x e^{itH_q} = M_x(t) = \begin{bmatrix} 0 & a_{12}e^{-itk_{12}} & \dots & 0 & 0 \\ a_{21}e^{itk_{21}} & 0 & \dots & 0 & 0 \\ \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & \dots & 0 & -a_{(d-1)d}e^{itk_{(d-1)d}} \\ 0 & 0 & \dots & -a_{d(d-1)}e^{-itk_{d(d-1)}} & 0 \end{bmatrix}$$

$$e^{-itH_q}M_y e^{itH_q} = M_y(t) = i\begin{bmatrix} 0 & a_{12}e^{-itk_{12}} & \dots & 0 & 0 \\ -a_{21}e^{itk_{21}} & 0 & \dots & 0 & 0 \\ \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & \dots & 0 & -a_{(d-1)d}e^{itk_{(d-1)d}} \\ 0 & 0 & \dots & a_{d(d-1)}e^{-itk_{d(d-1)}} & 0 \end{bmatrix}, \quad (2.8)$$

$$k_{ij} = k_{ji} = |I_{z,ii} + I_{z,jj}|, \quad a_{ij} = k_{ij}I_{x,ij}. \quad (2.9)$$

Both operators are antisymmetric relative to their secondary diagonals. In both matrices, only the off- diagonal elements closest to the main diagonals are nonzero. Combining operators (2.6) and (2.8) as well as I_x and I_y, we can construct a matrix that coincides with B_α^{m-n}, to within a coefficient. If the derived operator is taken as the effective Hamiltonian H_{eff}, then the evolution of the system under the action of H_{eff} during the time $t_d^{m-n}(\theta)$ will correspond to the selective rotation operator (2.4) between the corresponding pair of its levels through an angle θ. The phase difference between the states of various levels resulting from free evolution during the time $t_d^{m-n}(\theta)$ under the action of H_q plays a key role in the realization of selectivity. In this case, the constant q specifies a natural time and frequency scale. Therefore, in subsequent formulas, we will measure the time in units of $1/q$ and the frequency in units of q. Let us consider successively the cases with $d = 3$, 4, 5, and 6.

(1) $d = 3$. The effective Hamiltonians $H_{\text{eff}}t_3^{m-n}$ for the various qutrit rotations that correspond to the operator θB_α^{m-n} were obtained in [13]:

$$\theta B_x^{m-n} = (\pm M_x + I_x)t_3^{m-n}, \quad \theta B_y^{m-n} = (\mp M_y + I_y)t_3^{m-n}, \quad (2.10)$$

$$t_3^{m-n} = \frac{\theta}{2\sqrt{2}}.$$

The upper and lower signs in front of the operators correspond to the 0–1 and 1–2 transitions, respectively.

(2) $d = 4$. A feature of the half-integer spins 3/2 and 5/2 ($d = 4$ and 6) is the absence of matrix elements on the secondary diagonals in the operators M_α. Therefore, the rotation on the central transition should be obtained using a scheme slightly different from that described above. Let us first consider the two lateral transitions:

$$\theta B_x^{m-n} = (\pm M_x + M_y(\tau))t_4^{m-n}, \quad \theta B_y^{m-n} = (\mp M_y + M_x(\tau))t_4^{m-n}, \quad (2.11)$$

$$t_4^{0-1} = t_4^{2-3} = \frac{\theta}{4\sqrt{3}}, \quad \tau = \frac{\pi}{4}.$$

The upper and lower signs in front of the operators correspond to the 0–1 and 2–3 transitions, respectively.

The central transition can be obtained using the sum of operators ($\alpha = x, y$)

$$\theta B_\alpha^{1-2} = (I_\alpha + I_\alpha(\tau))t_4^{1-2}, \quad t_4^{1-2} = \frac{\theta}{4}, \quad \tau = \frac{\pi}{2}. \quad (2.12)$$

(3) $d = 5$. The effective Hamiltonian of the x rotation between states 0–1 and 3–4 is

$$\theta B_x^{m-n} = ([\pm M_x + I_x] + [-M_y(\tau) \pm I_y(\tau)])t_5^{m-n}, \quad (2.13a)$$

and between the states 1–2 and 2–3 is

$$\theta B_x^{m-n} = ([\pm M_x + 3I_x] - [-M_y(\tau) \pm 3I_y(\tau)])t_5^{m-n} \quad (2.13b)$$

(the upper and lower signs in front of the operators correspond to the 0–1 (1–2) and 3–4 (2–3) transitions, respectively). In Eqs. (2.13),

$$t_5^{0-1} = t_5^{3-4} = \frac{\theta}{16}, \quad t_5^{1-2} = t_5^{2-3} = \frac{\theta}{8\sqrt{6}}, \quad \tau = \frac{\pi}{2}.$$

For the y rotation, we should interchange the subscripts x and y and reverse the signs $\pm$ in the effective Hamiltonian (2.13).

(4) $d = 6$. The effective Hamiltonian of the x rotation between states 0–1 and 4–5 is

$$\begin{aligned}\theta B_x^{m-n} &= ([M_y(\tau_1) + M_y(\tau + \tau_1)] \\ &\pm [M_x + M_x(\tau)])t_6^{m-n},\end{aligned} \quad (2.14a)$$

and between states 1–2 and 3–4 is

$$\begin{aligned}\theta B_x^{m-n} &= ([M_y(2\tau_1) - M_y(\tau + 2\tau_1)] \\ &\pm [M_x - M_x(\tau)])t_6^{m-n}\end{aligned} \quad (2.14b)$$

(the upper and lower signs in front of the operators correspond to the 0–1 (1–2) and 4–5 (3–4) transitions, respectively). In Eqs. (2.14),

$$t_6^{0-1} = t_6^{4-5} = \frac{\theta}{16\sqrt{5}}, \quad t_6^{1-2} = t_6^{3-4} = \frac{\theta}{16\sqrt{2}},$$

$$\tau = \frac{\pi}{2}, \quad \tau_1 = \frac{\pi}{8}.$$

For the y rotation, we should interchange the subscripts x and y and reverse the sign $\pm$ in the effective Hamiltonian (2.14).

The central transition occurs under the operator ($\alpha = x, y$)

$$\begin{aligned}\theta B_\alpha^{2-3} &= ([I_\alpha + I_\alpha(\tau)] + [I_\alpha(\tau) + I_\alpha(\tau + \tau_1)])t_6^{2-3}, \\ t_6^{2-3} &= \frac{\theta}{12}, \quad \tau = \frac{\pi}{2}, \quad \tau_1 = \frac{\pi}{4}.\end{aligned} \quad (2.15)$$

3. CONSTRUCTING THE COMPOSITE SELECTIVE ROTATION OPERATOR USING NONSELECTIVE OPERATORS

In the previous section, we formally solved the problem of transforming H_q into the operator $H_{\text{eff}} = \theta B_\alpha^{m-n}/t_d^{m-n}$ represented as the sum $\sum_k H_k$. Let us now turn to making this transformation using the sequence of rotation operators (2.1) separated by intervals of free evolution with the Hamiltonian H_q. Let us substitute the representation of H_{eff} as the sum into the evolution operator

$$\exp(-iH_{\text{eff}}t) = \exp\left(-it\sum_k H_k\right). \quad (3.1)$$

Formally, the sought-for sequence can be obtained from Eq. (3.1) if we rewrite its right-hand side as the product of exponentials and use the property of exponential operators:

$$\begin{aligned}&\exp(e^{-i\psi I_\alpha}(-itH_q)e^{i\psi I_\alpha}) \\ &= e^{-i\psi I_\alpha}e^{-itH_q}e^{i\psi I_\alpha} = \{\psi\}_\alpha - t - \{\psi\}_{-\alpha}, \\ &\exp(e^{-itH_q}(-iM_\alpha)e^{itH_q}) \\ &= e^{-itH_q}e^{-iM_\alpha}e^{itH_q} = -t - e^{-iM_\alpha} - (-t) - .\end{aligned} \quad (3.2)$$

In what follows, we will write the free evolution operator during time t in the expressions for the pulse sequences as "$-t-$".

In fact, the operators in sum (3.1) do not commute with each other. Therefore, to obtain a pulse sequence whose action is equivalent to the evolution with the Hamiltonian H_{eff}, we will use the Trotter–Suzuki formula for exponential operators [18]:

$$\begin{aligned}&\exp\left(-it\sum_k H_k\right) \\ &= \left(\prod_k \exp\left(-\frac{iH_k t_k}{N}\right)\right)^N + O\left(\left(\frac{t}{N}\right)^2\right), \quad \sum_k t_k = t.\end{aligned} \quad (3.3)$$

To improve convergence, we will symmetrize the product of operators in this formula. As applied to multipulse NMR spectroscopy, these methods have been well studied and described in monographs (see, e.g., [14, 15]). Below, we consider the pulse sequences separately for $d = 3$, 4, 5, and 6.

Note that "reverse" free evolution operators are encountered in Eq. (3.2):

$$\exp(iH_q t). \quad (3.4)$$

To obtain this operator, we can extend the time in the permitted evolution operator by a period T:

$$\exp(-iH_q(T - t)). \quad (3.5)$$

$T = 2\pi$ for integer spins ($d = 3$, 5) and $T = \pi$ for half-integer ones ($d = 4$, 6). This significantly increases the total duration of the sequence. The reverse free evolution (3.4) can also be achieved by a different method—using the composite pulse (A.11). In this paper, we will not use it, since the sequences become more complex and the error increases.

(1) $d = 3$. To obtain the pulse sequence that performs a selective rotation on a qutrit, let us write out the operator M_α in (2.10) via the operators K_x and K_y and then symmetrize the exponential operators using the ABCBA scheme [18]:

$$\begin{aligned}&(e^{-i\theta A/2N}e^{-i\theta B/2N}e^{-i\theta C/N}e^{-i\theta B/2N}e^{-i\theta A/2N})^N \\ &= e^{-i\theta(A+B+C)} + O\left(\left(\frac{\theta}{N}\right)^3\right).\end{aligned} \quad (3.6)$$

For the y rotation, it is convenient to choose $A = K_x$, $B = K_y$, and $C = I_y$. As a result, we arrive at the pulse sequences obtained previously in [13]. For example, the selective y rotation between the 0–1 states of a qutrit can be written using (2.10) and (3.6) as the sequence of operators

$$\begin{aligned}&\Bigg[\left\{\frac{\pi}{4}\right\}_x - \frac{\theta}{2\sqrt{2}N} - \left\{\frac{\pi}{4}\right\}_{-x}\left\{\frac{\pi}{2}\right\}_y - \frac{\theta}{4\sqrt{2}N}\\ &-\left\{\frac{\theta}{2\sqrt{2}N}\right\}_{-y} - \frac{\theta}{4\sqrt{2}N} - \left\{\frac{\pi}{2}\right\}_{-y}\left\{\frac{\pi}{4}\right\}_x\\ &-\frac{\theta}{2\sqrt{2}N} - \left\{\frac{\pi}{4}\right\}_{-x}\Bigg]^N.\end{aligned} \tag{3.7}$$

(2) $d = 4$. When the pulse sequence is constructed for spin $I = 3/2$, it is important that the operators in sum (2.11) commute with each other (since there is no central coupling transition). Therefore, only the operators M_α rather than the entire sum need to be symmetrized. In this case, we will symmetrize the operators according to the ABA scheme:

$$\begin{aligned}&(e^{-i\theta A/2N}e^{-i\theta B/N}e^{-i\theta A/2N})^N\\ &= e^{-i\theta(A+B)} + O\left(\left(\frac{\theta}{N}\right)^3\right),\end{aligned} \tag{3.8}$$

where $A = K_x$ and $B = K_y$. Furthermore, operators (2.6) and (2.8) correspond to the simultaneous rotation on the two lateral transitions but in the opposite and the same directions, respectively. This can be used to obtain more complex gates.

For the y rotation on the 0–1 states, we obtain the sequence

$$\begin{aligned}&\Bigg[\left\{\frac{\pi}{4}\right\}_{-x} - \frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{4}\right\}_x\left\{\frac{\pi}{2}\right\}_{-y}\\ &-\frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{2}\right\}_y\left\{\frac{\pi}{4}\right\}_{-x} - \frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{4}\right\}_x\Bigg]^N - \frac{\pi}{4}\\ &-\Bigg[\left\{\frac{\pi}{4}\right\}_y - \frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{4}\right\}_{-y}\left\{\frac{\pi}{2}\right\}_x - \frac{\theta}{4\sqrt{3}N}\\ &-\left\{\frac{\pi}{2}\right\}_{-x}\left\{\frac{\pi}{4}\right\}_y - \frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{4}\right\}_{-y}\Bigg]^N - \frac{3\pi}{4} - .\end{aligned} \tag{3.9}$$

Using the commutative property of the z-rotation operators $\left\{\frac{\pi}{2}\right\}_y\left\{\frac{\pi}{4}\right\}_{\pm x}\left\{\frac{\pi}{2}\right\}_{-y}$ and $\left\{\frac{\pi}{2}\right\}_{-x}\left\{\frac{\pi}{4}\right\}_{\pm y}\left\{\frac{\pi}{2}\right\}_x$ with a quadrupole Hamiltonian, this sequence can be simplified:

$$\begin{aligned}&\Bigg[\left\{\frac{\pi}{4}\right\}_{-x} - \frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{2}\right\}_{-y}\\ &-\frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{2}\right\}_y - \frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{4}\right\}_x\Bigg]^N - \frac{\pi}{4}\\ &-\Bigg[\left\{\frac{\pi}{4}\right\}_y - \frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{2}\right\}_x - \frac{\theta}{4\sqrt{3}N}\\ &-\left\{\frac{\pi}{2}\right\}_{-x} - \frac{\theta}{4\sqrt{3}N} - \left\{\frac{\pi}{4}\right\}_{-y}\Bigg]^N - \frac{3\pi}{4} - .\end{aligned} \tag{3.10}$$

For the y rotation on the 1–2 states (central transition), we obtain the sequence

$$\left[\left\{\frac{\theta}{8N}\right\}_y - \frac{\pi}{2} - \left\{\frac{\theta}{4N}\right\}_y - \frac{\pi}{2} - \left\{\frac{\theta}{8N}\right\}_y\right]^N. \tag{3.11}$$

(3) $d = 5$. In sequence (3.3) of the effective Hamiltonian (2.13), we first symmetrize the terms in square brackets just as for a three-level system and then the entire sum according to the ABA scheme. For the y rotation on the 0-1 states, we obtain the sequence

$$\begin{aligned}&\left[\mathrm{Seq}_1 - \frac{\pi}{2} - \mathrm{Seq}_2 - \frac{3\pi}{2} - \mathrm{Seq}_1\right]^N,\\ &\mathrm{Seq}_1 \equiv \Bigg[\left\{\frac{\pi}{4}\right\}_{-x} - \frac{\theta}{32N^2} - \left\{\frac{\pi}{4}\right\}_x\left\{\frac{\pi}{2}\right\}_{-y}\\ &-\frac{\theta}{64N^2} - \left\{\frac{\theta}{32N^2}\right\}_y\\ &-\frac{\theta}{64N^2} - \left\{\frac{\pi}{2}\right\}_y\left\{\frac{\pi}{4}\right\}_{-x} - \frac{\theta}{32N^2} - \left\{\frac{\pi}{4}\right\}_x\Bigg]^N,\\ &\mathrm{Seq}_2 \equiv \Bigg[\left\{\frac{\pi}{4}\right\}_y - \frac{\theta}{16N^2} - \left\{\frac{\pi}{4}\right\}_{-y}\left\{\frac{\pi}{2}\right\}_x\\ &-\frac{\theta}{32N^2} - \left\{\frac{\theta}{16N^2}\right\}_{-x}\\ &-\frac{\theta}{32N^2} - \left\{\frac{\pi}{2}\right\}_{-x}\left\{\frac{\pi}{4}\right\}_y - \frac{\theta}{16N^2} - \left\{\frac{\pi}{4}\right\}_{-y}\Bigg]^N.\end{aligned} \tag{3.12}$$

(4) $d = 6$. Since the operators in square brackets in Eqs. (2.14) commute with each other, only the sum of

the operators inside the brackets and the operators M_α themselves should be symmetrized.

For the y rotation on the 0–1 states, we obtain the sequence

$$(\mathrm{Seq}_1)^N - \frac{\pi}{8} - (\mathrm{Seq}_2)^N - \frac{7\pi}{8},$$

$$\mathrm{Seq}_1 \equiv \left[\left\{\frac{\pi}{4}\right\}_{-x} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{-y} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{y} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{4}\right\}_{x}\right]^N - \frac{\pi}{2}$$
$$- \left[\left\{\frac{\pi}{4}\right\}_{-x} - \frac{\theta}{16\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{-y} - \frac{\theta}{16\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{y} - \frac{\theta}{16\sqrt{5}N^2} - \left\{\frac{\pi}{4}\right\}_{x}\right]^N$$
$$- \frac{\pi}{2} - \left[\left\{\frac{\pi}{4}\right\}_{-x} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{-y} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{y} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{4}\right\}_{x}\right]^N, \quad (3.13)$$

$$\mathrm{Seq}_2 \equiv \left[\left\{\frac{\pi}{4}\right\}_{y} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{x} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{-x} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{4}\right\}_{-y}\right]^N - \frac{\pi}{2}$$
$$- \left[\left\{\frac{\pi}{4}\right\}_{y} - \frac{\theta}{16\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{x} - \frac{\theta}{16\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{-x} - \frac{\theta}{16\sqrt{5}N^2} - \left\{\frac{\pi}{4}\right\}_{-y}\right]^N$$
$$- \frac{\pi}{2} - \left[\left\{\frac{\pi}{4}\right\}_{y} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{x} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{2}\right\}_{-x} - \frac{\theta}{32\sqrt{5}N^2} - \left\{\frac{\pi}{4}\right\}_{-y}\right]^N,$$

where we made a transformation similar to (3.10).

For the y rotation on the 2–3 states (central transition), we obtain the sequence

$$\mathrm{Seq}_1 - \frac{\pi}{4} - \mathrm{Seq}_1 - \frac{3\pi}{4},$$
$$\mathrm{Seq}_1 \equiv \left[\left\{\frac{\theta}{24N}\right\}_{y} - \frac{\pi}{2} - \left\{\frac{\theta}{12N}\right\}_{y} - \frac{\pi}{2} - \left\{\frac{\theta}{24N}\right\}_{y}\right]^N. \quad (3.14)$$

The sequences for the rotations about the x axis and other transitions can be obtained from the above ones after the substitution of operators that can be easily derived from the form of Eqs. (2.10)–(2.15).

4. NUMERICAL SIMULATION OF THE REALIZATION OF THE COMPOSITE SELECTIVE ROTATION OPERATOR USING NONSELECTIVE RF PULSES

To control a quadrupole nucleus, we will apply a variable RF magnetic field. An RF pulse is produced under the action of a field with amplitude B_1 and frequency ω for a finite time $t_p \gg 1/\omega$. We will consider rectangular RF pulses, i.e., we will assume that the RF field is switched on and off instantly, while its amplitude is constant during the entire pulse. In a reference frame rotating with frequency ω [17], the change of the state with time is specified by an evolution operator,

$$U(t) = e^{-iHt} \quad (4.1)$$

with a time-independent effective Hamiltonian,

$$H = (\omega - \omega_0)I_z + q\left(I_z^2 - \frac{1}{3}I(I+1)\right) + \Omega(I_x\cos\varphi + I_y\sin\varphi). \quad (4.2)$$

Here, $\omega_0 = \gamma B_0$ is the Larmor spin precession frequency and $\Omega = \gamma B_1$ is the RF field amplitude. The RF field phase φ defines the field direction in this reference frame. For rotation (2.1) to be realized, the RF field amplitude in Eq. (4.2) must be much larger than the difference between the resonance frequencies of various transitions, i.e., $\Omega \gg q$. Let us take $\omega = \omega_0$. For an RF pulse corresponding to (2.1), we then obtain from (4.1)

$$P_\alpha(\theta) = \exp(-it_p(H_q + \Omega I_\alpha)), \quad (4.3)$$

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

The formulas of the previous section hold rigorously when the ideal nonselective rotation operators (2.1) are used. In a real experiment, these operators can be obtained using the evolution operator (4.3). The presence of a quadrupole interaction simultaneously with the RF field leads to errors that disappear only in the limit $\Omega \longrightarrow \infty$ (see Appendix). In particular, since the sign of the quadrupole interaction does not change as the sign of the rotation angle (the RF field direction) changes, the condition

$$P_\alpha(\theta)P_{-\alpha}(\theta) = 1$$

and, as a result, Eqs. (3.2) do not hold.

Characteristics of the composite selective rotation operators $R_{\alpha}^{m-n}(\theta)$ for various d

m–n	$T_p = \Sigma t_p$	$T_c = \Sigma t_c$	T_∞	N_S
		$d = 3$		
0–1, 1–2	$\frac{\theta}{2\sqrt{2}\Omega} + \frac{2\pi N}{\Omega}$	$\frac{N}{\Omega}\left(4a\left(\frac{\pi}{4}\right) + 2a\left(\frac{\pi}{2}\right) + a\left(\frac{\theta}{2\sqrt{2}N}\right)\right)$	$\frac{3\theta}{2\sqrt{2}}$	$7N$
		$d = 4$		
0–1, 2–3	$\frac{3\pi N}{\Omega}$	$\frac{4N}{\Omega}\left(a\left(\frac{\pi}{4}\right) + a\left(\frac{\pi}{2}\right)\right)$	$\frac{3\theta}{2\sqrt{3}} + \pi$	$8N$
1–2	$\frac{\theta}{2\Omega}$	$\frac{N}{\Omega}\left(2a\left(\frac{\theta}{8N}\right) + a\left(\frac{\theta}{4N}\right)\right)$	πN	$3N$
		$d = 5$		
0–1, 3–4	$\frac{\theta}{8\Omega} + \frac{6\pi N^2}{\Omega}$	$\frac{N^2}{\Omega}\left(12a\left(\frac{\pi}{4}\right) + 6a\left(\frac{\pi}{2}\right) + 2a\left(\frac{\theta}{32N^2}\right) + a\left(\frac{\theta}{16N^2}\right)\right)$	$\frac{3\theta}{8} + 2\pi N$	$21N^2$
1–2, 2–3	$\frac{3\theta}{8\Omega} + \frac{6\pi N^2}{\Omega}$	$\frac{N^2}{\Omega}\left(12a\left(\frac{\pi}{4}\right) + 6a\left(\frac{\pi}{2}\right) + 2a\left(\frac{3\theta}{32N^2}\right) + a\left(\frac{3\theta}{16N^2}\right)\right)$	$\frac{3\theta}{4\sqrt{6}} + 2\pi N$	$21N^2$
		$d = 6$		
0–1, 4–5	$\frac{9\pi N^2}{\Omega}$	$\frac{12N^2}{\Omega}\left(a\left(\frac{\pi}{4}\right) + a\left(\frac{\pi}{2}\right)\right)$	$\frac{3\theta}{4\sqrt{5}} + \pi(2N+1)$	$24N^2$
1–2, 3–4	$\frac{9\pi N^2}{\Omega}$	$\frac{12N^2}{\Omega}\left(a\left(\frac{\pi}{4}\right) + a\left(\frac{\pi}{2}\right)\right)$	$\frac{3\theta}{4\sqrt{2}} + \pi(2N+1)$	$24N^2$
2–3	$\frac{\theta}{3\Omega}$	$\frac{2N}{\Omega}\left(2a\left(\frac{\theta}{24N}\right) + a\left(\frac{\theta}{12N}\right)\right)$	$\pi(2N+1)$	$6N$

Note: N_S is the number of nonselective rotation operators (simple or composite) in the sequence, $T_p = \Sigma t_p$ is the total duration of the simple nonselective RF pulses, $T_c = \Sigma t_c$ is the total duration of the composite nonselective RF pulses, and T_∞ is the total duration of the intervals of free evolution.

Another known method for reducing the error of nonselective rotation consists in using composite pulses [15, 16]. In particular, composite pulses were suggested in [16] to reduce the error of the rotation due to the quadrupole interaction for a nucleus with $I = 1$. Unfortunately, the suggested variants do not remove the phase distortions and, therefore, are inefficient for our purposes. In the Appendix, we obtained composite pulses that consist of five simple pulses and that remove the linear (in q/Ω) contribution to the error for arbitrary spins. In the general case, the sequence for the rotation through an angle θ around the x axis can be written as

$$P_{-y}\left(\frac{3\pi}{2}\right) - \tau_1 - P_{-y}\left(\frac{\pi}{2}\right)P_{-x}\left(\frac{3\pi}{2}\right) - \tau_2 - P_x(\psi_1)P_x(\psi_2), \tag{4.4}$$

$$b = \arcsin\left(\frac{\sqrt{2}}{2}\sin\theta\right), \quad \psi_1 = \frac{\pi}{2} - b, \quad \psi_2 = \theta - b,$$

where the times of free evolution are defined as

$$\begin{aligned}\tau_1 &= \frac{1}{\Omega}\left[\pi + \frac{1}{2}\left(\theta - 2b - \sin 2b + \frac{1}{2}\sin 2\theta\right)\right],\\ \tau_2 &= \frac{1}{\Omega}\left[\pi - \sin 2b + \frac{1}{2}\sin 2\theta\right].\end{aligned} \tag{4.5}$$

For the y rotation, the subscripts x and y should be interchanged.

In particular, for a nonselective rotation around the x axis through π/2 and π/4, we find in (4.4), respectively,

$$\begin{aligned}\psi_1 = \psi_2 = \frac{\pi}{4}, \quad \tau_1 &= \left(\pi - \frac{1}{2}\right)\frac{1}{\Omega},\\ \tau_2 &= \frac{\pi - 1}{\Omega},\end{aligned} \tag{4.6}$$

and

$$\psi_1 = \frac{\pi}{3}, \quad \psi_2 = \frac{\pi}{12},$$

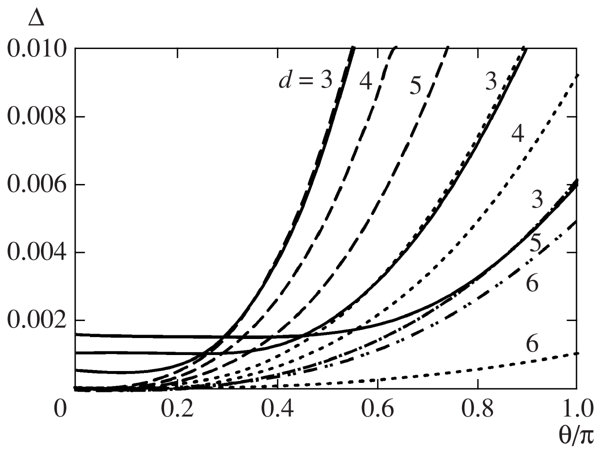

Fig. 1. Error of the composite selective rotations R_y^{0-1} versus angle θ for various d and various numbers of repetitions N in Eq. (3.3): $N = 1$ (dashed curves), $N = 2$ (dotted lines), and $N = 3$ (dash–dotted curves; only for $d = 3$). The solid curves indicate the error when composite nonselective RF pulses are used in the composite selective rotation operator $R_y^{0-1}(\theta)$ at $\Omega = 50q$ instead of the ideal rotation operators. The dotted curve for $d = 5$ and the dashed curve for $d = 6$ coincide.

$$\tau_1 = \left(\frac{23}{24}\pi - \frac{\sqrt{3}-1}{4}\right)\frac{1}{\Omega}, \tag{4.7}$$

$$\tau_2 = \left(\pi - \frac{\sqrt{3}-1}{2}\right)\frac{1}{\Omega}.$$

Thus, a nonselective spin rotation can be realized using a simple (4.3) or composite (4.4) RF pulse. Substituting these operators into the above formulas for composite selective rotations, we numerically simulated the experimental situation. The simulation results are presented in the figures for various selective rotations of various spins in the form of an error,

$$\Delta = \frac{1}{d}\sqrt{\sum_{i,j}\left|U_{ij} - U_{ij}^{\text{theor}}\right|^2}, \tag{4.8}$$

where U_{ij}^{theor} are the elements of the matrix of the ideal selective rotation operator $R_\alpha^{m-n}(\theta)$ (2.3) and U_{ij} are the elements of the numerically calculated matrix obtained using the product of evolution operators (4.1). Characteristics of the sequences of RF pulses are given in the table.

Figure 1 shows the results for the sequence of ideal nonselective rotation operators at various θ and I. This figure illustrates the angular dependences of the error resulting from the operators constituting H_{eff} being noncommutative. We see that this error increases with rotation angle. Therefore, we will have to divide the sequences into a larger number of cycles (to increase N) at large angles.

Such ideal rotations are achieved in the limit $\Omega/q \longrightarrow \infty$. As we see from Figs. 2 and 3, the error due to the distortion of the nonselective rotation operator (4.3) under the simultaneous action of the RF field and H_q is added at finite values of this parameter. This part of the error can be reduced by replacing the simple pulses in the sequences with composite ones (see Figs. 2 and 3 as well as Fig. 1 and the corresponding figure from [13]). Figure 2 presents the error of the selective rotation on spins $I = 1$ and 2. Figure 3 compares the errors of the selective rotations for various transitions of spin $I = 5/2$. A feature of the central tran-

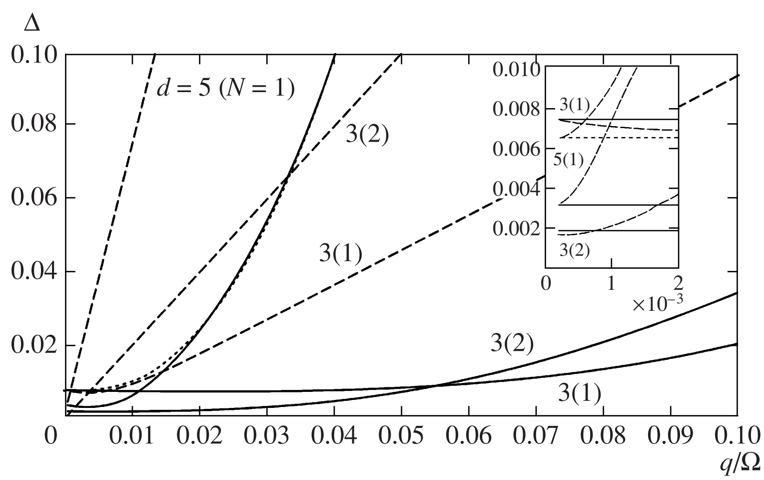

Fig. 2. Error of the composite selective rotations $R_y^{0-1}(\pi/2)$ and $R_y^{1-2}(\pi/2)$ for $d = 3$ and 5 versus reciprocal of the nonselective RF pulse amplitude. The solid and dashed lines indicate the dependences when, respectively, composite and simple nonselective RF pulses are used. The curves for both transitions at $d = 3$ coincide, while the 1–2 transition for $d = 5$ is highlighted by the dotted line. In the inset, the region near zero, where the error reaches the limiting value, is magnified.

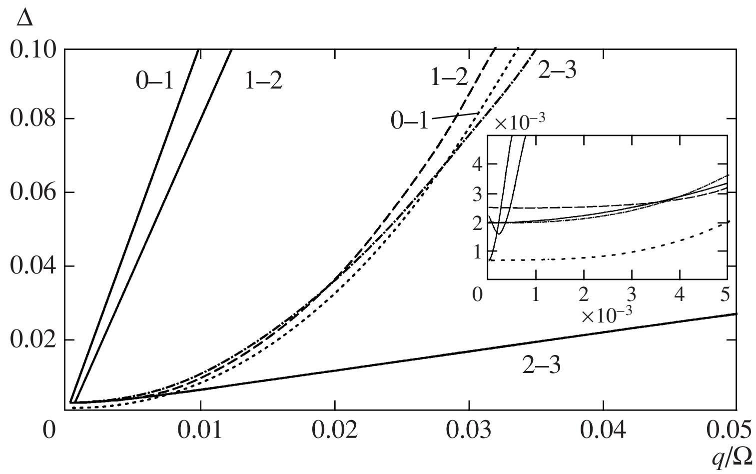

Fig. 3. Error of the composite selective rotations R_y^{m-n} ($\pi/2$) for $d = 6$ versus reciprocal of the nonselective RF pulse amplitude. The solid lines and other curves indicate the dependences when, respectively, simple and composite pulses are used. The numbers near the curves indicate the corresponding *m–n* transitions. In the inset, the region near zero, where the error reaches the limiting value, is magnified.

sition is a smaller error when using simple RF pulses in sequence (3.15) than that for composite nonselective rotations. The reason is easy to understand upon viewing Fig. 5 (see below). For the central transition, the RF pulse rotation angle of $\pi/24$ is small and falls to the left of the intersection between the lines for the dependences of the errors (i.e., simple pulses lead to a small errors; therefore, composite pulses increase it by adding the error from additional pulses). For the other two transitions in sequences (3.14), the angles are large ($\pi/4$ and $\pi/2$) and fall to the right of the intersection. Naturally, in the limit $\Omega \longrightarrow \infty$, the errors of the two sequences for each transition reach a common limiting value—the error for the sequence of ideal nonselective rotation operators. The same peculiarity is observed for the central transition of spin $I = 3/2$.

Figure 4 for $I = 3/2$ shows the dependence of the error on the durations of the selective ($\pi/2$) rotation operators, simple ($T_S = t_p = \theta/\Omega$) and composite ones, for the 0–1 and 1–2 transitions. The composite operator was obtained by two methods: first, using the sequence of simple nonselective pulses and, second, using the same sequence with composite nonselective rotation operators (4.4). The formulas for the durations of the sequences are given in the table. The corresponding pairs of curves converge as $T_S \longrightarrow T_\infty$, where T_∞ is the limiting value of the total duration defined by the total duration of the intervals of free evolution and is reached in the limit $\Omega \longrightarrow \infty$. $T_\infty = 1.433\pi$ for the 0–1 transition and $T_\infty = N\pi$ for the 1–2 transition, since the peculiarity of the central transition is an invariable duration of the intervals of free evolution $\tau = \pi/2$ as N increases. The minimum error Δ_∞ stems from the fact that the operators are noncommutative and decreases equally when passing from $N = 1$ to 2 for both transitions. For the 1–2 transition, the time T_∞ doubles.

At finite Ω, the duration of the sequences increases by the total duration of the RF pulses of simple, T_p, and composite, T_c, nonselective rotations (see table). The dependences shown in Fig. 4 can be understood by assuming that the RF pulse errors (A.7) or (A.8) (see Appendix) are added in the region $\Omega/q \longrightarrow \infty$ under consideration. For simple pulses,

$$\begin{aligned}\Delta_p &= \Delta_\infty + qI(I+1)\sum t_p f_p \\ &\approx \Delta_\infty + (T_S - T_\infty)b_p,\end{aligned} \tag{4.9}$$

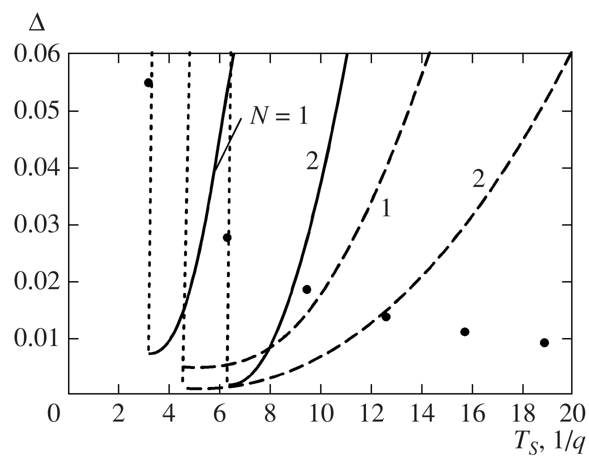

Fig. 4. Error of the realization of selective rotations R_y^{m-n} ($\pi/2$) for $d = 4$ versus RF pulse duration ($T_S = t_p$) and total duration of the pulse sequence. The values are shown for the 0–1 (dashed curves) and 1–2 (solid curves) transitions when composite nonselective pulses ($T_S = T_\infty + T_c$) are used. The dotted lines correspond to the sequences of simple nonselective pulses ($T_S = T_\infty + T_p$). For the rotation by a simple selective RF pulse, the dots indicate the minimum values of the rapidly oscillating error [13].

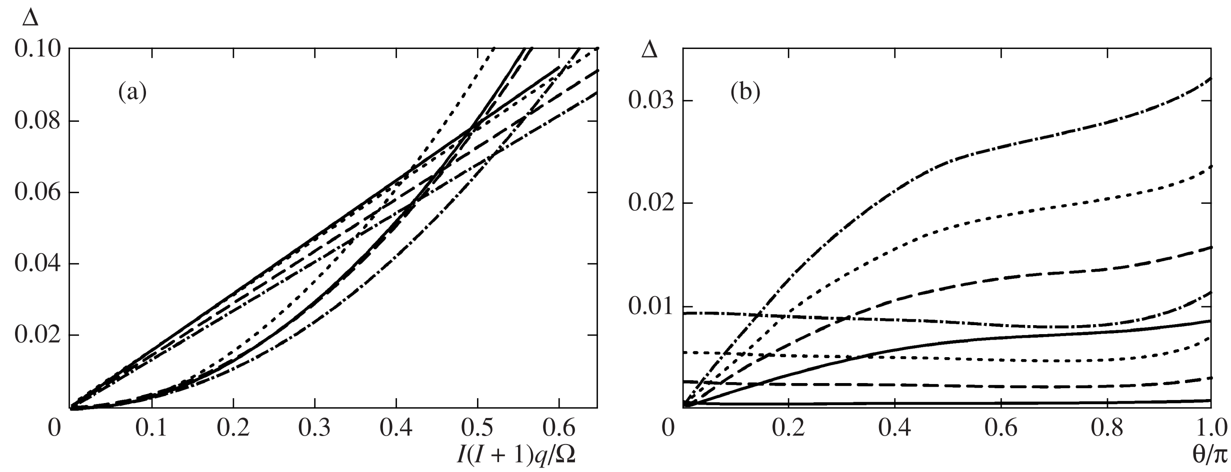

Fig. 5. Comparison of the errors of the realization of simple and composite nonselective y rotations: $d = 3$ (solid lines), 4 (dashed lines), 5 (dotted lines), and 6 (dash-dotted lines). (a) The dependences on the reciprocal of the RF pulse amplitude for a rotation angle of $\pi/2$. The parabolic curves and straight lines correspond to the composite and simple pulses, respectively. (b) The dependences on the angle θ at RF pulse amplitude $\Omega = 50q$ for simple (rising curves) or composite (weakly changing lines) pulses.

where b_p is a constant. Here, we approximately took f_p outside the sign of the sum and used the fact that $T_p = \sum t_p = T_S - T_\infty$. We see a linear dependence on T_S in Fig. 4. The parallelism of the straight dotted lines suggests that the corresponding coefficients are close. For composite nonselective RF pulses at fixed Ω, the duration and error of one such pulse depend weakly on the angle. Therefore, the total error is determined by their number N_S,

$$\begin{aligned}\Delta_c &= \Delta_\infty + q^2 I^2 (I+1)^2 \sum t_c^2 f_c \\ &\approx \Delta_\infty + (T_S - T_\infty)^2 b_c / N_S,\end{aligned} \tag{4.10}$$

since $t_c \approx (T_S - T_\infty)/N_S$. Here, b_c is a constant. The corresponding change in the shape of the parabolas with N_S is seen in Fig. 4.

For comparison, Fig. 4 shows the error (4.8) of the rotation due to an ordinary selective rectangular RF pulse. The dots indicate only the minimum values that are reached if the phase of the nonresonance levels change by 2π in $T_S = t_p = \theta/\Omega$. The error increases rapidly as the pulse duration changes (see [13]). We see that using composite selective rotation operators allows us to reduce the error for the same time (or to reduce the time for the same error). These properties are also observed in systems with a large number of levels.

As a specific example, let us turn to the experimental work [6], in which the qutrit states were controlled by NMR methods. A deuterium nucleus ($I = 1$) partially oriented in a liquid-crystalline matrix at room temperature was taken. The NMR spectrum of deuterium [6] consisted of two lines corresponding to $q = 120$ Hz in (2.5). The authors used both selective and nonselective RF pulses. For rectangular nonselective RF pulses, $\Omega/q \approx 100$. At these parameters, for a composite selective rotation through $\pi/2$ at $N = 1$ we find from the formulas given in the table that

$$\begin{aligned} T_\infty = 2.2\ \text{ms}, \quad T_\infty + T_p = 2.3\ \text{ms}, \\ T_\infty + T_c = 3.9\ \text{ms}. \end{aligned} \tag{4.11}$$

In [6], the duration of a simple Gaussian selective RF pulse for the rotation through $\pi/2$ was 6 ms. Although the duration of the composite selective pulse (4.11) at the parameters taken from the paper was shorter by only a factor of 1.5–2.0, the error of the operation will be significantly smaller, as can be inferred, for example, from a comparison of Fig. 2 in this paper with the results for a Gaussian pulse in Fig. 1 from [13].

In our calculations, we used rectangular RF pulses, which allowed the theoretical ideas to be demonstrated most clearly. Unfortunately, the RF pulses of real NMR spectrometers are never ideal, causing the error to increase as their number increases. To reduce such errors [15], pulses of a more complex shape or composite pulses are used in practice. Passing to them will not affect the qualitative conclusions of the suggested approach, although it will lead to complication of the formulas and calculations. Since these refinements are related to the characteristics of the specific instrument, the corresponding calculations have to be performed during the setup when the experiment is carried out.

In another paper [7], the same group of experimenters considered a quadrupole ^{23}Na nucleus ($I = 3/2$) with four levels in a liquid-crystalline matrix. In the number of RF pulses needed for a composite selective rotation, the case of four levels is no more complex than the previous one (see table) and even simpler for the central transition. However, in this case, a larger splitting of the

NMR spectrum is observed, since the quadrupole moment of sodium is larger than that of deuterium by almost a factor of 50. Therefore, conditions are more favorable for applying simple selective pulses. To provide the conditions for the application of composite selective pulses described above, we have to increase the RF field amplitude in comparison to the case of deuterium nuclei or to heat the sample to reduce the order parameter of the liquid crystal.

5. CONCLUSIONS

To reduce the error of an ordinary selective RF pulse, we have to reduce its amplitude and to increase its duration. At the chosen duration, $t_p \sim 1/q$, the error is finite. Changing the shape of a short pulse reduces it insignificantly [13]. Above, we showed that the error of the selective rotation could be reduced theoretically to zero if the rotation were performed using a sequence of intense nonselective RF pulses separated by intervals of free evolution under a quadrupole interaction. Because of the presence of such intervals, the total duration of the sequence T_S cannot be made smaller than some limiting value of T_∞ dependent on the quadrupole interaction (q and I) and on the arrangement of the sequence. In other words, for $T_S > T_\infty$, the theory allows the limit $\Delta \longrightarrow 0$ to be reached using composite selective pulses, while for a simple selective pulse $\Delta \longrightarrow 0$ only when $T_S \longrightarrow \infty$. The possibility of such a reduction in the error s important for quantum computing, since only at Δ smaller than some critical value can the error correction procedure be applied [1].

In conclusion, note that the derived sequences of nonselective rotation operators can be of use not only for the quadrupole nuclei considered above but also for the electron spins controlled by microwave or laser pulses in systems with weak axially symmetric spin–orbit coupling.

ACKNOWLEDGMENTS

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APPENDIX

Composite Nonselective Pulse

We will use a property of Hamiltonian (2.5) that follows from the property of the sum of the squares of three projection operators as the basis for constructing a composite pulse:

$$I_x^2 + I_y^2 + I_z^2 - I(I+1) = 0. \tag{A.1}$$

This explains the choice of two pairs of operators to compensate for the error at the ends of the intervals of free evolution in sequence (4.4). The fifth pulse is needed to produce the necessary combined rotation. Under the action of a rectangular pulse (4.3), the operator I_z changes not instantly but according to Eqs. (2.2), where $\theta = \Omega t$. Therefore, we obtain the more complex relations (4.5) for the parameters.

Thus, let a strong time-varying RF field (4.2) be applied to the magnetic moment of a nucleus. At $q = 0$, the evolution operator takes the form

$$\begin{aligned} U_0(t) \equiv \{\theta\}_\alpha = \hat{T}\exp\Bigg(i\int_0^t \Omega(\tau)[I_x\cos\varphi(\tau) \\ + I_y\sin\varphi(\tau)]d\tau\Bigg). \end{aligned} \tag{A.2}$$

At $q \neq 0$, by restricting ourselves to the first order in small quantity q/Ω, we obtain

$$U(t) = U_0(t)\left[1 - i\int_0^t U_0^{-1}(\tau)H_qU_0(\tau)d\tau\right]. \tag{A.3}$$

Based on (2.2), we will represent the time dependence of the magnetic moment operator as

$$U_0^{-1}(\tau)I_zU_0(\tau) = \mu_x(\tau)I_x + \mu_y(\tau)I_y + \mu_z(\tau)I_z. \tag{A.4}$$

After the substitution of this expression into Eq. (A.3), we obtain a system of six equations from the condition of the integral in parentheses being equal to zero:

$$\frac{1}{t}\int_0^t \mu_\alpha^2(\tau)d\tau = \frac{1}{3}, \quad \alpha = x, y, z, \tag{A.5}$$

$$\begin{aligned} \int_0^t \mu_x(\tau)\mu_z(\tau)d\tau = \int_0^t \mu_x(\tau)\mu_y(\tau)d\tau \\ = \int_0^t \mu_y(\tau)\mu_z(\tau)d\tau = 0. \end{aligned} \tag{A.6}$$

It is easy to verify that the composite pulse (4.4) satisfies these equations. Condition (A.2) is met for any b, since at $q = 0$ the ideal rotations

$$P_{-x}(\psi_1) = \left\{\frac{\pi}{2} - b\right\}_{-x}, \quad P_x(\psi_2) = \{\theta - b\}_x$$

are performed in opposite directions about the same axis. The value of this parameter can be determined from the last equality in (A.6). The positive contribution from the last two pulses to the integral should offset the negative contribution from the third pulse. The satisfaction of condition (A.5) is then achieved by choosing the duration of the intervals of free evolution τ_1 and τ_2.

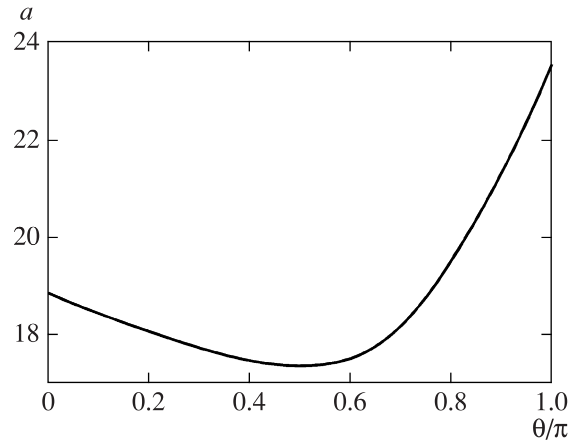

Fig. 6. Parameter a, which defines the duration of a composite nonselective RF pulse in Eq. (A.9), versus angle θ.

The dependences of error (4.8) on q/Ω and θ for simple and composite nonselective RF pulses are shown in Fig. 5 for various spins. The q/Ω dependence is linear for a simple pulse and quadratic for a composite one. The θ dependence of Δ is nearly linear for a simple pulse and is almost completely absent for a composite one. In addition, the error increases with I, since the quadrupole interaction becomes stronger. Qualitatively, the dependences of the errors on these parameters can be expressed by the following formulas for simple and composite pulses, respectively:

$$\Delta_p = qt_pI(I+1)f_p, \tag{A.7}$$

$$\Delta_c = q^2t_c^2I^2(I+1)^2f_c, \tag{A.8}$$

where f_p and f_c are nearly constant functions, which are nearly independent of Ω at $\Omega/I(I+1) > 5q$ and depend weakly (about 10%) on d and θ. Thus, for example, at $\theta = \pi/2$ and when d changes from 3 to 6, the function f_c is 1.11×10^{-3}, 1.33×10^{-3}, 1.10×10^{-3}, and 0.9×10^{-3}, while the function f_p is 0.1, 0.099, 0.093, and 0.087. The error increases with pulse durations t_p and t_c, which are inversely proportional to the RF field amplitude:

$$t_p = \theta/\Omega, \quad t_c = a(\theta)/\Omega. \tag{A.9}$$

The dependence $a(\theta)$ is defined by Eqs. (4.4) and (4.5) and is shown in Fig. 6. Whereas t_p depends linearly on θ, the dependence of t_c is much weaker, since the durations of two $3\pi/2$ pulses and one $\pi/2$ pulse make the main contribution to its value.

Relation (A.1) allows the reversal of the sign in front of the quadrupole interaction in the effective Hamiltonian to be achieved. For this purpose, let us rewrite it as

$$-q\left[I_z^2 - \frac{I(I+1)}{3}\right] = q\left[I_x^2 + I_y^2 - \frac{2I(I+1)}{3}\right]$$

and, accordingly, change conditions (A.5). In this case, substituting $\psi_1 = \pi/2$, $\psi_2 = 0$, and

$$\tau_2 = \tau_1 = t + \frac{\pi}{\Omega}, \tag{A.10}$$

into Eq. (4.4), we can obtain $-tH_q$.

To reduce the error, let us symmetrize the sequence using the well-known narrowing sequence WHH-4 [14] as an example,

$$P_x\left(\frac{\pi}{2}\right) - \tau_1 - P_y\left(\frac{3\pi}{2}\right) - \tau_2 - P_y\left(\frac{\pi}{2}\right) - \tau_1 - P_x\left(\frac{3\pi}{2}\right), \tag{A.11}$$

where $2\tau_1 = t - \pi/\Omega$ and $\tau_2 = t$. Here, to remove the error from the finite duration, we used two $3\pi/2$ pulses instead of the identical increase in the rotation angle of all pulses to $\beta > \pi/2$ proposed in [14]. Our approach allows universal conditions for τ_1 and τ_2 to be obtained instead of the need for solving a transcendental equation for β every time when t and Ω change. Other sequences were also suggested to reverse the time [15, 19]. At a sufficiently large amplitude Ω, the contribution to the error arises from $(I_x)^2$ and $(I_y)^2$ being noncommutative at $I > 1$. This error can be removed using the Trotter–Suzuki formula by dividing t into N segments.

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