Quantum state tomography of large nuclear spins in a semiconductor quantum well: Robustness against errors as quantified by condition numbers

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We discuss methods of quantum state tomography for solid-state systems with a large nuclear spin. They are based on quantum well. Due to quadrupolar interactions, the Zeeman levels of these nuclear-spin devices become nonequidistant, forming a controllable four-level quantum system (known as quartit or ququart). The occupation of these levels can be selectively and coherently manipulated by multiphoton transitions using the techniques of nuclear magnetic resonance (NMR) [Yusa et al., Nature (London) 434, 101 (2005)]. These methods are based on an unconventional approach to NMR, where the longitudinal magnetization $M_z$ is directly measured. This is in contrast to the standard NMR experiments and tomographic methods, where the transverse magnetization $M_x$ is detected. The robustness against errors in the measured data is analyzed by using condition numbers. We propose several methods with optimized sets of rotations. The optimization is applied to decrease the number of NMR readouts and to improve the robustness against errors, as quantified by condition numbers. An example of state reconstruction, using Monte Carlo methods, is presented. Tomographic methods for quadrupolar nuclei with higher-spin numbers (including $I = 7/2$) are also described.

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I. INTRODUCTION

Quantum state tomography (QST) is a method for reconstruction of a quantum state in a series of measurements performed on an ensemble of identical quantum states. Quantum state engineering provides methods for synthesis of quantum states, their coherent control and characterization. The latter task can be realized by quantum state and process tomographic methods.

Quantum state tomography (QST) is a method for reconstruction of a quantum state in a series of measurements performed on an ensemble of identical quantum states. Quantum process tomography (QPT) is a method, closely related to QST, which enables a complete characterization of the dynamics of a quantum system. Both QST and QPT have been applied widely to QIP in finite- and infinite-dimensional optical systems (for reviews see Refs. [2] [3] and references therein). In particular, much work has been done on QST of polarization states of photons (see, e.g., Refs. [4] [5]), homodyne QST [9], and homodyne QPT [10] [11] probed with coherent states. Other examples include QST in superconducting circuits [12] [13]. QST has also been applied in nuclear-spin systems using nuclear magnetic resonance (NMR) spectroscopy, which was motivated by quantum information interest [14] [15]. The NMR QST and NMR QPT were first developed for liquid-state nuclear spin-1/2 systems [15], and only later applied both to liquid- and solid-state systems of quadrupolar nuclei of spin-3/2 [16] [19] and spin-7/2 [20].

The use of multi-level systems (so-called qudits) instead of two-level systems (qubits) is an alternative paradigm [21] [22] in QIP, which has attracted attention in recent years. Standard examples of higher-order nuclear spins include: $I = 3/2$ for the isotopes $^{69}$Ga, $^{71}$Ga, and $^{73}$As (in, e.g., GaAs), $I = 5/2$ for $^{27}$Al (in, e.g., AlN) or $^{121}$Sb (in, e.g., FeSb$_2$), $I = 7/2$ for $^{123}$Sb (also in FeSb$_2$), as well as $I = 9/2$ for $^{113}$In and $^{115}$In (in, e.g., InAs, InSb, and InP), and $^{73}$Ge. Note large nuclear spins occur also in molecular magnets, which use clusters of spins, which can be applied for QIP [23] [24]. As another example, the superconducting circuits in Ref. [21] have up to five levels and can model rotations of spin-1 and spin-3/2. Here, we will focus on QIP using quadrupolar nuclei with spin-3/2 and spin-7/2, which are equivalent to four-level and eight-level systems, respectively.

Another motivation for the application of qudits for QIP is related to an important question concerning the scalability of two qubits to many qubits. If one simply plans to increase the number $N$ of qubits, then the re-
quired numbers of levels scales up exponentially. This becomes very hard to implement when $N \to \infty$. However, some ordinary classical computers are not assembled with simple AND, NAND, OR, NOT gates, but instead they are constructed using higher-level logic gates. Similarly, quantum computers might be constructed with slightly more complex logic gates, rather than, e.g., only Hadamard and CNOT gates. In this direction, multi-level systems are helpful.

A word of caution: replacing qubits by qudits causes a faster exponential divergence in the number of levels, thus one loses the advantage of “using a single multi-level system” over “using many two-level systems.” Thus, this approach should be applied carefully. Indeed, using qudits could reduce the complexity of quantum computers (see Ref. [21] and the justifications given there). Moreover, for qudits, there are optimal recipes for gate operations (see Sec. III) and quantum tomography methods to be discussed in the following sections.

Various two-qubit quantum state engineering methods and quantum algorithms have been realized in NMR experiments with spin-3/2 systems. Examples include: the demonstration of classical [25,27] and quantum [10,17,28,29] gates, generation of Bell states [28,29], the quantum Fourier transform [17], and implementations of simple quantum algorithms (i.e., the two-qubit Grover search algorithm [17,30] and the Deutsch-Jozsa algorithm [17,31]). The existence of quantum correlations (as revealed by quantum discord) was also experimentally demonstrated in spin-3/2 systems (see, e.g., [22]). It is worth noting that a prerequisite for the realization of all these gates and algorithms is the preparation of pseudo pure states (see also Refs. [14,33,34] and references therein).

Quadrupolar nuclei with spin-7/2 have also attracted increasing interest, as it is highly desirable to scale QIP beyond two (real or virtual) qubits. A few NMR experiments were performed with spin-7/2 systems, e.g., the preparation of effective pure states [35], a quantum simulation [36], a half-adder and subtractor operations [27,37], a test of phase coherence in electromagnetically-induced transparency [38], or three-qubit Deutsch-Jozsa algorithms [20,39,40].

A complete verification of the generated states and/or performed algorithms in the aforementioned experiments requires the application of QST.

In this article, we describe QST methods for an unconventional approach to NMR (sometimes referred to as exotic NMR) in semiconductor nanostructures [33,41–44], which is based on the measurement of the longitudinal magnetization $M_z$.

In contrast to this approach, the vast majority of the NMR tomographic methods are based on conventional (standard) NMR experiments, where the transverse magnetization $M_{xy}$ is detected. Indeed, a very tiny magnetic field produced by the nuclear spin rotation in the $xy$ plane with a resonant frequency is picked up by a surrounding coil. In this method, the $M_{xy}$ component is measured by using induction detection ($M_{xy}$ detection). However, this widely used conventional NMR suffers from low sensitivity arising from induction detection, so that one should prepare large volume samples occasionally reaching a cubic centimeter (at least a cubic millimeter). In the application to semiconductor (solid-state) systems (see Ref. [45] and references therein), multiple-layer quantum wells with 10-100 layers should be prepared to detect clear signals with a sufficient noise-to-signal ratio. A main advantage of semiconductor (solid-state) qubits is its precise controllability by using gate operations. Such gate operation is based on a single quantum well and nanostructure so that conventional NMR is obviously not appropriate for these systems. In the last decade, highly sensitive NMR methods suitable for semiconductor hetero- and nanosystems have been developed by using electrical [41,42] and optical [46] means. However, they all relied on a direct measurement of the nuclear spin magnetization, i.e., $M_z$ detection. Therefore, it is important for semiconductor (solid-state) nuclear-spin qubits to develop QST appropriate for the direct detection of $M_z$.

In Sec. II, we specify the quadrupolar interaction model. In Sec. III, we describe sequences of NMR pulses for implementing qubit gates in qudits. In Sec. IV, we present the key aspects of the $M_z$-based QST of a spin-3/2 system and specify the observation approaches to be applied in the next sections. We also briefly discuss a nanometer-scale all-electrical resistively-detected NMR device [41,42], where the $M_z$-magnetization can be measured. In Sec. V, we describe the linear reconstruction of density matrices in relation to condition numbers describing how these methods are robust against errors. In Sec. VI, we describe simple sets of rotations which enable the $M_z$-based QST of a spin-3/2 system. In Sec. VII, we show how to construct sets of operations optimized as regards the number of NMR readouts. We also find single-photon replacements for multi-photon rotations. In Sec. VIII, we describe generalized QST methods for higher-spin systems including spin-7/2. The results of our numerical simulations and application of the maximum-likelihood method are presented in Sec. IX. In Appendices, for completeness and clarity, we define selective rotations, and shortly compare the $M_{xy}$ and $M_z$ detections.

II. INTERACTION MODEL

First, we describe a model for large nuclear spins in a semiconductor quantum well and interacting with radiofrequency (RF) pulses. A general description of such interaction can be found in standard textbooks on NMR (see, e.g., Refs. [47,48]). In particular, the model described in detail by Leuenberger et al. [49], which was directly applied in the experiment of Yusa et al. [42], can be also adapted here.

Specifically, we analyze an ensemble of quadrupolar
nuclei (with spin $I > 1/2$) in a semiconductor quantum well interacting with $N$ RF pulses of the carrier frequency $\omega^{(k)}$, phase $\phi^{(k)}$, and magnetic-field amplitude $B_k$ ($k = 1, 2, ..., N$) in the presence of a strong magnetic field $B_0$. The effective total Hamiltonian in the laboratory frame reads

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}},$$

being a sum of the free term

$$\mathcal{H}_0 = \mathcal{H}_Z + \mathcal{H}_Q = \hbar \omega_0 I_z + \frac{\hbar \omega_Q}{3} (3I_z^2 - I^2),$$

and the term describing the interaction of the nuclei with

$$\mathcal{H}_{\text{int}} = \sum_{k=1}^{N} \frac{\hbar \omega_k}{2} \left[ I_z e^{-i(\omega^{(k)} t + \phi^{(k)})} + I_z e^{i(\omega^{(k)} t + \phi^{(k)})} \right]$$

$$= \sum_{k=1}^{N} \hbar \omega_k \left[ I_z \cos(\omega^{(k)} t + \phi^{(k)}) + I_y \sin(\omega^{(k)} t + \phi^{(k)}) \right].$$

Here, $\mathcal{H}_Z = \hbar \omega_0 I_z$ and $\mathcal{H}_Q$ describe, respectively, the Zeeman and quadrupole splittings (see Fig. 1 in the special case for spin $I = 3/2$). The operator $I_z$ (for $\alpha = x, y, z$) is the $\alpha$-component of the spin angular momentum operator, and $I_{\pm} = I_x \pm iI_y$. Moreover, $\omega_0 = -\gamma B_0$ is the nuclear Larmor frequency, and $\omega_k = -\gamma B_k$ is the amplitude (strength) of the $k$th pulse, where $\gamma$ is the gyromagnetic ratio. For the example of the nuclei $^{60}$Ga and $^{71}$As of spin $I = 3/2$ in semiconductor GaAs, we can choose the gyromagnetic ratios to be $\gamma(^{60}\text{Ga}) = 1.17 \times 10^7 \text{s}^{-1}\text{T}^{-1}$ and $\gamma(^{71}\text{As}) = 7.32 \times 10^6 \text{s}^{-1}\text{T}^{-1}$, which are estimated from the spectra measured in Ref. [42]. The Hamiltonian $\mathcal{H}_Q = \mathcal{H}_Q^{(1)} + \mathcal{H}_Q^{(2)} + ...$ describes the quadrupolar interaction as a sum of the first- and second-order quadrupolar terms (as shown in Fig. 1 for spin $I = 3/2$), but also higher-order terms. The first-order quadrupolar splitting parameter (quadrupolar frequency) $2\omega_Q$ is given for solids by [50]:

$$\omega_Q \equiv \omega_Q^{(1)} = \frac{3\pi C_Q}{4T(2T - 1)} (3\cos^2 \theta_Q - 1),$$

where $C_Q$ is the quadrupolar coupling constant, and $\theta_Q$ is the angle between the direction of the field $B_0$ and the principle axis of the electric-field gradient tensor. We assume a uniaxial electric-field gradient tensor, i.e., the biaxiality parameter is zero ($\eta_Q = 0$). Under the secular approximation, which is valid for relatively small $\omega_Q$, the effective interaction is described solely by the first-order quadrupolar Hamiltonian, as we have assumed in Eq. (2).

The quadrupolar frequencies are typically of the order of 10–100 kHz. For example, the values for the isotopes in semiconductor GaAs can be found in Refs. [42] [49] [51]. In our numerical simulations, we set the following values of the quadrupolar frequencies $\omega_Q(^{60}\text{Ga}) = 15.2 \text{kHz}$ and $\omega_Q(^{71}\text{As}) = 26.9 \text{kHz}$. These values were estimated from the experimental spectra reported in Ref. [42]. Moreover, we also choose in our simulations the same values of parameters as those measured or estimated in the experiment with nanometer-scale device in Ref. [42]. Namely, $B_0 = 6.3 \text{T}$ and $B_k = 0.2–1.4 \text{mT}$, and decoherence time is $T_2 \approx 1 \text{ms}$. Let us denote the eigenvalues and eigenvectors of $\mathcal{H}_0$ by $\epsilon_m$ and $|m\rangle$, respectively, i.e., $\mathcal{H}_0 |m\rangle = \epsilon_m |m\rangle$. If the condition $|\epsilon_m| \ll |\omega_Q| \ll |\omega_0|$ is satisfied, one can apply a selective RF pulse resonant with a transition $|m\rangle \leftrightarrow |n\rangle$, i.e., $\hbar \omega_{\text{RF}} = \epsilon_m - \epsilon_n$, where $m, n = 0, 1, ...$. One can also analyze $N$-photon resonant transitions, which correspond to the condition $N \omega_{\text{RF}} = \epsilon_m - \epsilon_n$, where $k = 1, 2, ...$
The evolution of a state, given by \( \rho \), is described in the rotating frame by:

\[
\rho(t) = U(\omega_1, t) \rho(t) U^\dagger(\omega_1, t),
\]

where the evolution operator is

\[
U(\omega_1, t) = \exp[-(i/\hbar)\mathcal{H}_{\text{rot}} t],
\]

The evolution of \( \rho(t) \) in the absence of pulses from the time \( t \) to \( t + \Delta t \) is given by:

\[
\rho(t + \Delta t) = U(0, \Delta t) \rho(t) U^\dagger(0, \Delta t).
\]

Analytical expressions for the evolution operator \( U(\omega_1, t) \) and the corresponding density matrices can be obtained by finding eigenvalues and eigenstates of \( \mathcal{H}_{\text{rot}} \). For example, by assuming an RF pulse to be resonant with the central line (i.e., \( \omega_{R1} = \omega_{12} = \omega_0 \)), we find the following eigenvalues of \( \mathcal{H}_{\text{rot}} \):

\[
eig(\mathcal{H}_{\text{rot}}) = \left[ \frac{\omega_1}{2} + \Omega_{\pm}, \frac{\omega_1}{2} - \Omega_{\pm}, -\frac{\omega_1}{2} - \Omega_{+}, -\frac{\omega_1}{2} + \Omega_{+} \right]
\]

where \( \Omega_{\pm} = \sqrt{\omega_1^2 \pm \omega_1 \Omega_Q + \omega_1^2} \). The corresponding eigenvectors of \( \mathcal{H}_{\text{rot}} \) for \( m = 1, 2 \) and \( n = 3, 4 \) are equal to:

\[
|V_m\rangle = \mathcal{N}_m [\sqrt{3} \omega_1 (|3\rangle + |0\rangle)] + y_m (|1\rangle + |2\rangle)], \]

\[
|V_n\rangle = \mathcal{N}_n [\sqrt{3} \omega_1 (|3\rangle - |0\rangle)] + z_n (|1\rangle - |2\rangle)],
\]

where \( \mathcal{N}_m \) and \( \mathcal{N}_n \) are normalization constants, and

\[
y_m = \omega_1 + 2\Omega_{\pm} + (-1)^m \omega_Q, \]

\[
z_n = \omega_1 + (-1)^n \Omega_{+} + 2\omega_Q.\]

The general solution for \( U(\omega_1, t) \) is quite lengthy. However, by assuming that \( |\omega_1| \gg |\omega_Q| \gg |\omega_1| \), it can be effectively reduced to a form corresponding to all ideal selective rotations as defined in Appendix A. This can be shown by expanding the elements of the matrix \( U(\omega_1, t) \) in a power series of the parameter \( \epsilon = |\omega_1|/|\omega_Q| \), and finally keeping only the first term of this expansion.

For example, if the pulse is resonant with the central transition, then the evolution operator \( U(\omega_1, t) \) can be approximated by

\[
U_{12}(\omega_1, t_p) = \begin{bmatrix}
\delta^* & 0 & 0 & 0 \\
\delta \cos(\omega_1 t_p) & -i\delta \sin(\omega_1 t_p) & 0 & 0 \\
0 & -i\delta \sin(\omega_1 t_p) & \delta \cos(\omega_1 t_p) & 0 \\
0 & 0 & 0 & \delta^*
\end{bmatrix},
\]

where \( \delta = \exp(i\omega_Q t_p) \). It is seen that \( U_{12}(\omega_1, t) \) reduces to the perfect selective rotation \( X_{12}(\theta) = R^{(X)}_{12}(\theta) \), with \( \theta = 2\omega_1 t_p \), if the pulse duration is chosen such that \( \omega_Q t_p \) is a multiple of \( 2\pi \). Analogously, other rotations \( R^{(i)}_{mn}(\theta) \), given by Eq. (A4), can be implemented for \( i = X, Y, Z \) and \( m, n = 0, \ldots, 3 \) with \( m \neq n \).

III. IMPLEMENTING GATES IN MULTI-LEVEL SPIN SYSTEMS

Here, we discuss how to implement single- and two-qubit gates in systems with spins 3/2 and 7/2. This can enable formally simple implementations of arbitrary multi-qubit quantum algorithms by applying sequences of NMR pulses in multi-level spin systems.

A. Spin-3/2 system

We focus on various NMR QST methods for a system with spin-3/2 nuclei. Due to the Zeeman and quadrupo-
lar interactions (shown in Fig. 1), this system is described in an external magnetic field by a non-equidistant four-level energy spectrum. Thus, this spin-3/2 system can be referred to as a quartit (also called ququart or four-level qudit). The basic set of eigenfunctions of the system can be described with the states $|m\rangle_A|n\rangle_B$ of two logical (or virtual) qubits $A$ and $B$ corresponding to an ensemble of identical spin-1/2 pairs:

$$
\begin{align*}
|\frac{3}{2}, \frac{3}{2}\rangle &\equiv |0\rangle \equiv |00\rangle, \quad |\frac{3}{2}, -\frac{3}{2}\rangle \equiv |2\rangle \equiv |10\rangle, \\
|\frac{3}{2}, \frac{1}{2}\rangle &\equiv |1\rangle \equiv |01\rangle, \quad |\frac{3}{2}, -\frac{1}{2}\rangle \equiv |3\rangle \equiv |11\rangle.
\end{align*}
$$

(16)

A pure state of a quartit can be written in this basis states as

$$
|\psi\rangle = c_0|0\rangle + c_1|1\rangle + c_2|2\rangle + c_3|3\rangle
$$

(17)
in terms of the normalized complex amplitudes $c_i$, so an arbitrary mixed state of a quartit is described by a density matrix $\rho = [\rho_{nm}]_{4\times4}$. 

Our discussion in this section is based on a fundamental theorem in quantum information according to which any quantum gate can be constructed from single-qubit rotations and any nontrivial two-qubit gate, e.g., the CNOT gate. In a quartit, rotations of a virtual qubit $A$ and $B$, denoted respectively by $R^A(\theta)$ and $R^B(\theta)$, can be implemented by the application of two pulses:

$$
\begin{align*}
R^A(\theta) &= R_{02}(\theta)R_{13}(\theta), \\
R^B(\theta) &= R_{01}(\theta)R_{23}(\theta),
\end{align*}
$$

(18)

where $R_{mn}(\theta)$ (with $R = X, Y, Z$) is a selective rotation resonant with a transition between levels $|m\rangle$ and $|n\rangle$ as defined in Appendix A (see also Fig. 2).

Note that realizations of single virtual qubit gates in a qudit are more complicated than those for real qubits. In contrast to those, usually virtual qubit gates can be realized much simply, e.g., a CNOT-like gate can be implemented by applying a single $\pi$-pulse, e.g.,

$$
U'_{\text{CNOT}} \equiv U'_{\text{CNOT12}} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \end{bmatrix} = Y_{23}(\pi).
$$

(19)

Similarly, a SWAP-like gate can also be implemented easily by a single $\pi$-pulse:

$$
U'_{\text{SWAP}} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} = Y_{12}(\pi).
$$

(20)

The above CNOT-like and SWAP-like gates can be related to the standard CNOT and SWAP gates as follows:

$$
U_{\text{CNOT}} = SU'_{\text{CNOT}}, \quad U_{\text{SWAP}} = U'_{\text{SWAP}}S,
$$

(21)

where $S = \text{diag}([1, 1, -1, 1])$. It is worth noting that any unitary operator that can create entanglement between a pair of qubits (or virtual qubits) is universal. Thus, the standard SWAP gate $U_{\text{SWAP}}$ is not universal, as its entangling power is zero. In contrast to this gate, the SWAP-like gate $U'_{\text{SWAP}}$ is universal, as it can entangle qubits.

\section*{B. Spin-7/2 system}

As a generalization of the tomographic methods of spin-3/2 nuclei, we now analyze a spin-7/2 nuclei, e.g., for the isotope $^{123}$Sb in the semiconductor FeSb$_2$. We neglect the case of a spin-5/2 nuclei (corresponding to a six-level qudit) for which not all three-qubit states can be encoded. For this reason such system is much less interesting for the standard QIP. Obviously, the tomography of spin-5/2 nuclei can still be easily applied.

A spin-7/2 system in an external magnetic field $B_0$ corresponds to a quoctit (eight-level qudit), which is equivalent to three logical qubits (e.g., labeled as $A, B$ and $C$). So, its basis states can be identified with the three-qubit states:

$$
\begin{align*}
|\frac{7}{2}, \frac{7}{2}\rangle &\equiv |0\rangle \equiv |000\rangle, \quad |\frac{7}{2}, -\frac{7}{2}\rangle \equiv |4\rangle \equiv |100\rangle, \\
|\frac{7}{2}, \frac{5}{2}\rangle &\equiv |1\rangle \equiv |001\rangle, \quad |\frac{7}{2}, -\frac{5}{2}\rangle \equiv |5\rangle \equiv |101\rangle, \\
|\frac{7}{2}, \frac{3}{2}\rangle &\equiv |2\rangle \equiv |010\rangle, \quad |\frac{7}{2}, -\frac{3}{2}\rangle \equiv |6\rangle \equiv |110\rangle, \\
|\frac{7}{2}, \frac{1}{2}\rangle &\equiv |3\rangle \equiv |011\rangle, \quad |\frac{7}{2}, -\frac{1}{2}\rangle \equiv |7\rangle \equiv |111\rangle.
\end{align*}
$$

(22)

where $|klm\rangle \equiv |k\rangle_A|l\rangle_B|m\rangle_C$. Any pure state of such system can be described by $|\psi\rangle = \sum_{k,l,m=0}^7 c_k |k\rangle$, in the basis spanned by states given by Eq. (22). QST of spin-7/2 systems corresponds to a reconstruction of a mixed state described by a density matrix $\rho = [\rho_{nm}]_{8\times8}$.

Analogously to a quartit, we find that virtual qubits in a quoctit can be selectively rotated by applying combinations of four pulses:

$$
\begin{align*}
R^A(\theta) &= R_{04}(\theta)R_{15}(\theta)R_{26}(\theta)R_{37}(\theta), \\
R^B(\theta) &= R_{02}(\theta)R_{13}(\theta)R_{46}(\theta)R_{57}(\theta), \\
R^C(\theta) &= R_{01}(\theta)R_{23}(\theta)R_{45}(\theta)R_{57}(\theta).
\end{align*}
$$

(23)

CNOT-like gates, i.e., $U'_{\text{CNOT}kl}$ with $k$ ($l$) denoting a control (controlled) bit, can be obtained by combinations of two pulses:

$$
\begin{align*}
U'_{\text{CNOT}12} &= Y_{46}(\theta)Y_{57}(\theta), \\
U'_{\text{CNOT}13} &= Y_{45}(\theta)Y_{67}(\theta), \\
U'_{\text{CNOT}23} &= Y_{23}(\theta)Y_{67}(\theta).
\end{align*}
$$

(24)

Analogously, SWAP-like gates in quoctit can be implemented as follows:

$$
\begin{align*}
U'_{\text{SWAP}12} &= Y_{24}(\theta)Y_{35}(\theta), \\
U'_{\text{SWAP}13} &= Y_{14}(\theta)Y_{36}(\theta), \\
U'_{\text{SWAP}23} &= Y_{12}(\theta)Y_{56}(\theta).
\end{align*}
$$

(25)
From Eqs. \([24]\) and \([25]\), it is seen that realizations of CNOT and SWAP gates in a qudit are more complicated than those in a qudit, but still simpler than the standard CNOT and SWAP implementations in two real qudits.

It is worth noting that implementations of double controlled rotations are the simplest in a qudit in comparison to other dimensional systems. For example, the Toffoli-like (CCNOT-like) gates can be implemented by just a single \(\pi\)-pulse:

\[
\begin{align*}
U'_{123} &= Y_{67}(\pi), \\
U'_{132} &= Y_{57}(\pi), \\
U'_{231} &= Y_{37}(\pi),
\end{align*}
\]

(26)
where \(k\) and \(l\) \((m)\) in \(U'_{klm}\) denote control \((\text{controlled})\) bits.

IV. PRINCIPLES OF \(M_z\)-BASED QST

NMR quantum state tomography is a method for the complete reconstruction of a given density matrix \(\rho\) in a series of NMR measurements. In general, to completely reconstruct a density matrix \(\rho\) for a qudit or two qudits, we need to determine 15 real parameters, while the 16th element can be found from the normalization condition.

Single NMR readout can only give some elements of \(\rho\): either diagonal \((\text{in case of } M_z\text{-detection})\) or off-diagonal elements \((\text{for } M_{xy}\text{-detection})\), as discussed in Appendix B. The remaining matrix elements of the original density matrix \(\rho\) can be obtained by rotating it through properly chosen rotational operations \(R^{(k)}\), which change \(\rho\) as follows:

\[
\rho^{(k)} = R^{(k)} \rho (R^{(k)})^\dagger.
\]

(27)
These operations are performed before NMR readout measurements. Thus, the reconstruction of a given density matrix is possible by transforming \(\rho\) through various rotations \(R^{(k)}\) in such a way that all elements go over into measurable ones in a given detection method.

In the standard NMR \(M_{xy}\)-detection, one can directly determine some off-diagonal elements of the density matrix as given by Eqs. \([B1]\) and \([B2]\) in Appendix B. In contrast to the \(M_{xy}\)-detection, one directly determines only diagonal elements in the \(M_z\)-detection. It is worth noting that the spectrum of a spin-3/2 system obtained via the \(M_z\)-detection contains less information than the spectra obtained by the \(M_{xy}\)-detection as discussed in Appendix B: the \(M_{xy}\)-detection of a spin-3/2 system yields six real values, which correspond to three peaks of real and those of imaginary parts of the spectrum. Note that the \(M_{xy}\)-detection of a coupled two spin-1/2 system yields four values or even eight values if one could detect signals from ensembles of two different spins simultaneously.

Tomography based on the measurements of the \(M_z\) and \(M_{xy}\) magnetizations of spin-3/2 systems has been performed in experiments reported in Refs. \([16]\) and \([17]\), respectively.

A. Observation approaches

In an ideal \(M_z\)-detection, one can directly access all diagonal elements

\[
b_n^{(k)} = \rho_{nn}^{(k)}
\]

(28)
of any rotated density matrix \(\rho^{(k)} = R^{(k)} \rho (R^{(k)})^\dagger\) for \(k = 1, \ldots, N_r\), where \(N_r\) is the number of readouts \((\text{operations or sets of rotations})\). We refer to this purely theoretical approach as the observation approach \(I\), and the resulting coefficient matrices \(A'\) and \(C'\), defined in Sec. V, are marked with primes.

However, in a real experimental approach, referred to as the observation approach \(II\), the information is gathered from \(M_z\)-spectra, where one can roughly estimate the population differences \((\rho_{11} - \rho_{00}, \rho_{22} - \rho_{11}, \text{and } \rho_{33} - \rho_{22})\) from the amplitude of the signals by integrating the area of the peaks centered at \(\omega_{01}, \omega_{12}, \text{and } \omega_{23}\), respectively. Thus, on including the normalization condition, we have the following set of equations:

\[
b_n^{(k)} = N_r^{-1} (\rho_{n+1,n+1}^{(k)} - \rho_{nn}^{(k)}) = N_r^{-1} \text{Tr} (I_z^{(n+1,n)} \rho^{(k)}),
\]

(29)
for each rotated density matrix \(\rho^{(k)}\), where \(I_z^{(n+1,n)} = [n + 1\langle n + 1\rangle - n\langle n\rangle]_{\{\}} \) is the fictitious spin-1/2 operator for general spin and the constant \(N_r\) is usually chosen so that the thermal equilibrium magnetization vector is equal to a unit vector along the \(z\)-axis \([50]\).

The set of equations \([29]\) directly leads to the coefficient matrices \(A''\) and \(C''\), which are in general different from \(A'\) and \(C'\), obtained from Eq. \([28]\). Nevertheless, from Eq. \([29]\), one can easily determine all diagonal elements \(\rho^{(k)}:\)

\[
\begin{align*}
\rho_{00}^{(k)} &= \frac{1}{4} - \frac{1}{4} N (3b_1^{(k)} + 2b_2^{(k)} + b_3^{(k)}), \\
\rho_{11}^{(k)} &= \frac{1}{4} + \frac{1}{4} N (b_1^{(k)} - 2b_2^{(k)} - b_3^{(k)}), \\
\rho_{22}^{(k)} &= \frac{1}{4} + \frac{1}{4} N (b_1^{(k)} + 2b_2^{(k)} - b_3^{(k)}), \\
\rho_{33}^{(k)} &= \frac{1}{4} + \frac{1}{4} N (b_1^{(k)} + 2b_2^{(k)} + 3b_3^{(k)}).
\end{align*}
\]

(30)
In fact, the measured resistance in experiments performed in, e.g., Refs. \([33\) \([32\) \([33]\), is proportional to the longitudinal magnetization \(M_z \propto \text{Tr}[\rho I_z]\) defined in terms of the total angular momentum operator \(I_z = \text{diag}(\frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2})\) for spin \(I = 3/2\):

\[
M_z^{(k)} \propto \text{Tr}[\rho^{(k)} I_z] = \frac{N_r}{2} (3\rho_{00}^{(k)} + \rho_{11}^{(k)} - \rho_{22}^{(k)} - 3\rho_{33}^{(k)}).
\]

(31)
Here, we study a measurement method by applying the standard cyclically ordered phase sequence (CYCLOPS) to a \(\pi/20\) reading pulse and receiver \([52]\). The method was used in, e.g., an experiment on QST for quadrupolar nuclei of a liquid crystal by Bonk et al. \([16]\). In this
approach, NMR spectra were obtained from free induction decay (FID) averaged over each phase \((x, -y, -x, y)\). This enables the suppression of receiver imperfections and thus the cancellation of artifacts from the NMR spectra. The intensities \(b_n^{(k)}\) of three \((n = 1, 2, 3)\) peaks of the averaged NMR spectrum for rotated deviation matrices \(\Delta \rho^{(k)} = \rho^{(k)} - I/4\) \((k = 1, \ldots, N)\) together with the normalization conditions are described by the following set of equations for, e.g., the quartet:

\[
[b_1^{(k)}, b_2^{(k)}, b_3^{(k)}]^T = V \text{diag}(\Delta \rho^{(k)}),
\]

\[
0 = \text{Tr}(\Delta \rho^{(k)}),
\]

where \(V\):

\[
V = \begin{bmatrix}
\sqrt{3} e_{11} e_{12} & -\sqrt{3} e_{12} e_{22} & -\sqrt{3} e_{23} e_{13} & -\sqrt{3} e_{13} e_{14} \\
2 e_{13} e_{12} & 2 e_{22} e_{23} & -2 e_{23} e_{22} & -2 e_{13} e_{12} \\
\sqrt{3} e_{13} e_{14} & \sqrt{3} e_{13} e_{23} & \sqrt{3} e_{12} e_{22} & -\sqrt{3} e_{11} e_{12} \\
\end{bmatrix}
\]

The \(n\)th NMR peak corresponds to the transition between levels \(|n - 1\rangle\) and \(|n\rangle\). Above, \(\text{diag}(\Delta \rho^{(k)})\) denotes a column vector of the diagonal elements of \(\Delta \rho^{(k)}\). The coefficients \(e_{ij}\) are the absolute values of the \(\pi/20\) hard-reading pulse given by:

\[
[e_{ij}] = \frac{1}{4} \begin{bmatrix}
    c_{31} & s_{zz} & c_{z,-z} & s_{3,-1} \\
    s_{zz} & c_{13} & s_{-1,3} & c_{z,-z} \\
    c_{z,-z} & c_{-1,3} & c_{13} & s_{zz} \\
    s_{3,-1} & c_{z,-z} & s_{zz} & c_{31} \\
\end{bmatrix}
\]

where \(c_{xy} = x \cos \left(\frac{\pi}{40}\right) + y \cos \left(\frac{3\pi}{40}\right)\), \(s_{xy} = x \sin \left(\frac{\pi}{40}\right) + y \sin \left(\frac{3\pi}{40}\right)\), and \(z = \sqrt{3}\). We refer to this approach, given by Eqs. \(32\), as the observation approach III. It is clear that this method results in the coefficient matrices \(A''\) and \(C'''\) different from those obtained from Eqs. \(28\) and \(29\).

B. An implementation of \(M_z\)-detection in a nanometer-scale device

Here, we shortly describe an implementation of the NMR detection of the longitudinal magnetization of a small ensemble of quadrupolar spins 3/2, which is beyond the detection limits of conventional NMR techniques.\[41,43,46\]

This NMR detection was developed and applied in Refs. \[42\] to an on-chip semiconductor device based on a quantum-well structure shown in Fig. 3. This nanometer-scale device is composed of a monolithic GaAs quantum well integrated with a point contact channel and an antenna gate, where an RF field can be locally applied. The GaAs layer effectively forms a two-dimensional electron gas. The point-contact channel is composed of isotopes \({}^{69}\text{Ga}, {}^{71}\text{Ga}, \text{and } {}^{75}\text{As}\) having total ground-state spin \(I=3/2\). The nuclear spins in the channel can be selectively polarized by flowing current, while the spins in the other regions are kept in thermal equilibrium. These interactions between electron and nuclear spins are enhanced when an external static magnetic field \(B_0\) is applied to set the system at the spin phase transition of the Landau level filling factor 2/3.\[43\] The polarization is followed by RF pulses applied through the antenna gate, which enable manipulation of the nuclear spins. This coherent manipulation results in oscillations of the resistance of the point-contact channel, which are directly related to the oscillations in the longitudinal magnetization \(M_z\). Reference \[42\] observed clear oscillations reflecting all possible transitions between the four nuclear spin states (see Fig. 2) of each nuclide (\({}^{69}\text{Ga}, {}^{71}\text{Ga}, \text{and } {}^{75}\text{As}\)). This novel device, exhibiting extremely low deco-
herence \[12\ [44\), opens new perspectives to study char-
characteristics of nuclear spins in nanoscale semiconductors,
but also to precisely control nuclear spin states. The ar-
bitrary control of the superposition of the four spin-3/2
states enables the implementation of two-qubit coherent
operations.\[33\ [43\). Thus, the device offers new possi-
bilities to perform single- and two-qubit quantum gates,
or even to test simple quantum-information processing
algorithms. A fabrication of analogous device based on
InAs and InSb \[53\), instead of GaAs, where the isotopes
\[113\ In and \[115\ In have spin \(j = 9/2\) (a 10-level qudit) would
enable the implementation of three-qubit quantum gates
and algorithms. But it must be admitted that the de-
vices are not easily scalable for much higher number of
virtual and/or real qubits.

The initialization of the described device is relatively
easy. We can realize the effective pure state \[3\] by using
current-induced nuclear spin polarization with randomiz-
ing pulses of \(\omega_0\) and \(\omega_1\). Once the state \[3\] is realized, it
is transferred to \([0], [1],\) and \([2]\) by applying a respective
\(\pi\) pulse as described by us in Refs. \[33\ [39\).

The estimated polarization of the nuclear spins is quite
high. Therefore, obtaining initial states with high purity
should be practically quite simple — at least to start with
the pseudo-pure states \([00]\) or \([11]\).

There are many different possibilities to measure spec-
tra. The following is the simplest example applied in
experiments described in Ref. \[33\). After the prepara-
tion of a desired state, we apply a pulse with a duration
of \(\pi\)-pulse and measure how a resistance
changes as a function of frequency. In case of the con-
tion of a desired state, we apply a pulse with a duration
experiments described in Ref. \[33\]. After the prepara-
tra. The following is the simplest example applied in

Various numerical procedures for reconstructing an un-
known density matrix \(\rho\) from experimental data have
been developed (see, e.g., Refs. \[2\ [3\ and references
therein). The simplest and most intuitive QST is based
on the linear inversion of a linear-system problem,
\(Ax = b,\) (35)

where the vector \(x = \text{vec}(\rho)\) corresponds to the state \(\rho\)
to be reconstructed. This vector can be defined in various
ways, e.g., for a quartit state as
\[x = \text{vec}(\rho) = [\rho_{00}, \text{Re}\rho_{01}, \text{Im}\rho_{01}, \text{Re}\rho_{02}, \text{Im}\rho_{02}, \ldots, \rho_{33}]^T,\] (36)

where \(\rho_{ij}\), for \(i \leq j\), are only included. Moreover, in
Eq. (35), \(b\) is the observation vector, which contains the
measured data; and \(A\) is the coefficient matrix, which
is also referred to as the rotation matrix, or the data
matrix in a more mathematical context. Thus, the el-
ment \(A_{ij}\) is the coefficient of \(x_i\) in the \(j\)th equation
\((j = 1, ..., N_{eqs})\) for a chosen measurement rotation. In
our context, the observation vector \(b_j\) corresponds to the
integrated area of the NMR spectra. The number \(N_{eqs}\)
of equations is given by \(N_r \times N_{eqs}\), assuming \(N_r\) readouts
(for a given measurement), where each of them yields
\(N_{eqs}\) values corresponding, e.g., to the number of peaks
of an NMR spectrum (including both real and imaginary
parts). Usually, an extra equation is added for the nor-
malization condition, \(\text{Tr}b = 1\). Thus, for a quartit, the
observation vector has \(N_{eqs}\) elements and the coefficient
matrix \(A\) is of dimensions \(N_{eqs} \times 16\).

Usually, there are more equations than unknowns.
Such over determined problems can be solved as
\[Cx = \tilde{b},\] (37)

where \(C = [C_{ij}]_{16 \times 16} = A^\dagger A\) and \(\tilde{b} = [b_j]_{16 \times 1} = A^\dagger b\). Equation (37) results from the standard least-squares-
fitting analysis based on the minimalization of
\[\chi^2 = ||Ax - \tilde{b}||^2.\] Thus, one can easily calculate the solution
\(x = C^{-1}\tilde{b}\) and, finally, reconstruct the sought density
matrix \(\rho = \text{vec}^{-1}(x)\).

Dozens of different linear-inversion-based QST proto-
cols have been proposed and applied (see, e.g., \[2\ [3\ and
references citing those). Then the question arises: which
of them are preferable for certain goals and tasks.

As an indicator of the quality of a linear-inversion-
based QST method, or more precisely its error robust-
ness (or error sensitivity), one can apply the so-called
condition number defined as \[55\ [57:\]
\[\text{cond}_{\alpha,\beta}(C) = \|C\|_{\alpha,\beta} ||C^{-1}||_{\beta,\alpha} \geq 1,\] (38)

where \(C\) is a nonsingular square matrix and the con-
vention is used that \(\text{cond}_{\alpha,\beta}(C) = +\infty\) for a singular
matrix \(C\). Moreover, \(\| \cdot \|_{\alpha,\beta}\) denotes the subordinate
matrix norm, which can be defined via the vector norms:
\[||C||_{\alpha,\beta} = \max_{x \neq 0} ||Cx||_{\beta} / ||x||_{\alpha}.\] Clearly, the condition
numbers depend on the applied norm. Here, we apply
the spectral and Frobenius norms.

The spectral norm (also refereed to as the 2-norm) is
given by the largest singular value of \(C\).

The Frobenius norm is
\[\|C\|_F = \left(\sum_{i=1}^{N_{eqs}} \sigma_i^2\right)^{1/2},\] which is given in terms of the singular
values \(\sigma_i\), leads to the condition number
\[\kappa_F(C) \equiv \text{cond}_F(C) = ||C||_F \|C^{-1}||_F.\] (40)
There are various geometrical, algebraic, and physical interpretations of condition numbers (see Ref. [8] and references therein, in addition to Refs. [55, 59]). In particular, according to the Gantmacher-Kahan theorem, the inverse of a condition number corresponds to the relative distance of a nonsingular square matrix $C$ to the set of singular matrices. Another, more physical interpretation can be given as follows [55]: Let us assume errors $\delta b$ in the observation vector $\tilde{b}$ which cause errors $\delta x$ in the reconstructed vector $x$:

$$C(x + \delta x) = \tilde{b} + \delta b,$$

then the following inequalities hold

$$\frac{1}{\operatorname{cond}_{_{\alpha, \beta}}(C)} \frac{||\delta b||}{||b||} \leq \frac{||\delta x||}{||x||} \leq \operatorname{cond}_{_{\alpha, \beta}}(C) \frac{||\delta b||}{||b||}.$$  \hspace{1cm} (42)

It is clearly seen that when a condition number $\operatorname{cond}_{_{\alpha, \beta}}(C) \approx 1$, then small relative changes in the observation vector $\tilde{b}$ cause equally small relative changes in the reconstructed state $x$. This interpretation can be generalized to include also errors $\delta C$ in the coefficient matrix $C$.

Thus, by applying this general theorem, given in Eq. (42), to QST, we can conclude that if $\operatorname{cond}_{_{\alpha, \beta}}(C)$ is small (large), then the coefficient matrix $C$ is well-conditioned (ill-conditioned), which means that the method is robust (sensitive) to errors in the observation vector $\tilde{b}$. For ill-conditioned QST, even a minor error in $\tilde{b}$ can cause a large error in $x$. Some instructive numerical examples of ill-conditioned problems are given in Refs. [8, 55].

Condition numbers were applied to estimate the quality of optical tomographic reconstructions in, e.g., Refs. [8, 29]. However, to our knowledge, these parameters have not been applied yet to analyze the quality of NMR QST. Below we propose a few NMR QST protocols and compare their error robustness based on the condition numbers (as summarized in Table I).

Note that the smallest singular value (or, equivalently, eigenvalue) $\sigma_{\min}(C) = \min[\text{svd}(C)] = ||C^{-1}||_2$ of $C$ is also sometimes used as an error-robustness parameter. This approach was applied in the analysis of an NMR QST method in, e.g., Ref. [60]. Although condition numbers are much better parameters of the error robustness, in comparison to $\sigma_{\min}(C)$, we have also showed explicitly the latter values in Table I for all the studied QST methods.

### VI. SIMPLE SETS OF ROTATIONS FOR THE $M_z$-BASED QST

In the following, we describe a few QST methods based on the $M_z$-detection and analyze their error robustness.

### TABLE I: Error robustness parameters for linear reconstructions of states of nuclear spins $I = \frac{3}{2}$ and $\frac{7}{2}$. Key: OA denotes the “Observation Approach”, * indicates that the corresponding method includes an additional identity operation, $R^{(0)} = I$. It is seen that the inclusion of this identity operation only slightly changes the condition numbers.

| Spin method rotations | OA | $\sigma_{\min}$ | $\sigma_{\max}$ | $\kappa_{\text{II}}$ | $\kappa_{\text{F}}$ |
|-----------------------|----|----------------|----------------|----------------------|------------------|
| $3/2$                 | 1  | Eq. (43)       | $1.27$         | $28.5$               | $532.9$          |
|                       | I  | 2              | $12$           | $6$                  | $34.24$          |
|                       | I* | 2              | $12$           | $6$                  | $34.24$          |
|                       | II | 1.71           | $48$           | $28.14$              | $68.99$          |
|                       | II*| 1.74           | $52$           | $29.87$              | $74.38$          |
|                       | III| 0.1            | $48$           | $487$                | $927$            |
|                       | III*| 0.1           | $52$           | $521$                | $994$            |
|                       | 2  | Eq. (52)       | $1.17$         | $28$                 | $20.39$          |
|                       | I  | 2              | $7$            | $3.5$                | $20.39$          |
|                       | I* | 2              | $7$            | $3.5$                | $20.39$          |
|                       | II | 1.17           | $24$           | $20.49$              | $56.48$          |
|                       | II*| 1.17           | $28$           | $23.90$              | $57.92$          |
|                       | III| 0.08           | $24$           | $295$                | $665$            |
|                       | III*| 0.08          | $28$           | $345$                | $682$            |
|                       | 3  | Eq. (53)       | $1.28$         | $52$                 | $22.64$          |
|                       | I  | 2              | $7$            | $3.5$                | $20.39$          |
|                       | I* | 2              | $7$            | $3.5$                | $20.39$          |
|                       | II | 1.17           | $24$           | $20.49$              | $51.14$          |
|                       | II*| 1.37           | $28$           | $20.47$              | $43.79$          |
|                       | III| 0.08           | $24$           | $295$                | $607$            |
|                       | III*| 0.10          | $28$           | $284$                | $536$            |
|                       | 4  | Eq. (56)       | $1.28$         | $456$                | $522.4$          |
|                       | I  | 2              | $7$            | $3.5$                | $20.39$          |
|                       | I* | 2              | $7$            | $3.5$                | $20.39$          |
|                       | II | 1.17           | $24$           | $20.49$              | $51.14$          |
|                       | II*| 1.37           | $28$           | $20.47$              | $43.79$          |
|                       | III| 0.08           | $24$           | $295$                | $607$            |
|                       | III*| 0.10          | $28$           | $284$                | $536$            |
|                       | 5  | Eq. (57)       | $1.37$         | $52$                 | $22.64$          |
|                       | I  | 2              | $57$           | $28.5$               | $532.9$          |
|                       | I* | 2              | $57$           | $28.5$               | $532.9$          |
|                       | II | 1.27           | $448$          | $354$                | $1197$           |
|                       | II*| 1.28           | $456$          | $356$                | $1218$           |
|                       | 6  | Eq. (58)       | $1.28$         | $456$                | $522.4$          |
|                       | I  | 2              | $15$           | $7.5$                | $105.9$          |
|                       | I* | 2              | $15$           | $7.5$                | $105.9$          |
|                       | II | 0.38           | $112$          | $291.5$              | $636.1$          |
|                       | II*| 0.40           | $120$          | $296.6$              | $661.8$          |

### QST method 1

Our first proposal for QST of a spin-$3/2$ system, is based on a natural choice of 12 rotations:

$$(R^{(k)})_k = \{Y_{01}, X_{01}, Y_{12}, X_{12}, Y_{23}, X_{23},$$
$$Y_{02}, X_{02}, Y_{13}, X_{13}, Y_{03}, X_{03}\},$$  \hspace{1cm} (43)

where hereafter $X_{mn} \equiv X_{mn}(\frac{\pi}{2})$ and $Y_{mn} \equiv Y_{mn}(\frac{\pi}{2})$ as special cases of the selective rotations $X_{mn}(\theta)$ and $Y_{mn}(\theta)$ defined in Appendix A. Thus, the method is based on 6 single-photon, 4 two-photon, and 2 three-
 photon transitions. The $M_z$-based tomography for the rotations, given by Eq. (43), can be understood as follows: When we apply the $Y_{01}$ pulse after a certain photon operation, we obtain the diagonal components including $\rho_{01}$ and $\rho_{10}$. In our $M_z$-detection method, one of the three signals, which corresponds to the differences of the populations between four spin states, is proportional to $\text{Re}(\rho_{01}) + \text{Re}(\rho_{10})$. Because $\rho_{mn} = \rho_{nm}^*$, we can obtain $\text{Re}(\rho_{01}) = \text{Re}(\rho_{10})$. Similarly, $Y_{12}$, $Y_{23}$, $Y_{02}$, $Y_{13}$, and $Y_{03}$ give us other elements $\text{Re}(\rho_{mn}) = \text{Re}(\rho_{nm})$. Imaginary parts are also estimated by applying the $X_{mn}$ pulse by noting that $\text{Im}(\rho_{mn}) = -\text{Im}(\rho_{nm})$. Although QST needs $6 \times 2$ measurements and two (three)-photon operations, this method looks simple and easy to interpret.

In the $M_z$-detection approach, we can determine the diagonal elements $[\rho_{00}, \rho_{11}, \rho_{22}, \rho_{33}]$ of a density matrix $\rho$. By denoting the diagonal elements of $\rho^{(k)}$ as $\text{diag}(\rho^{(k)}) = (\rho_{nn})^k$, the following elements:

\[
\begin{align*}
\text{diag}(\rho^{(1)}) &= \{f^{(22)}_{00}, f^{(00)}_{01}, f^{(00)}_{02}, \rho_{33}\}, \\
\text{diag}(\rho^{(2)}) &= \{f^{(13)}_{01}, f^{(11)}_{01}, f^{(33)}_{01}, \rho_{33}\}, \\
\text{diag}(\rho^{(3)}) &= \{f^{(22)}_{12}, f^{(22)}_{12}, \rho_{33}\}, \\
\text{diag}(\rho^{(4)}) &= \{f^{(13)}_{12}, f^{(11)}_{12}, \rho_{33}\}, \\
\text{diag}(\rho^{(5)}) &= \{f^{(22)}_{23}, f^{(22)}_{23}, \rho_{33}\}, \\
\text{diag}(\rho^{(6)}) &= \{f^{(13)}_{23}, f^{(11)}_{23}, \rho_{33}\}, \\
\text{diag}(\rho^{(7)}) &= \{f^{(22)}_{02}, f^{(22)}_{02}, \rho_{33}\}, \\
\text{diag}(\rho^{(8)}) &= \{f^{(13)}_{02}, f^{(11)}_{02}, \rho_{33}\}, \\
\text{diag}(\rho^{(9)}) &= \{f^{(22)}_{12}, f^{(22)}_{12}, \rho_{33}\}, \\
\text{diag}(\rho^{(10)}) &= \{f^{(13)}_{12}, f^{(11)}_{12}, \rho_{33}\}, \\
\text{diag}(\rho^{(11)}) &= \{f^{(22)}_{03}, f^{(22)}_{03}, \rho_{33}\}, \\
\text{diag}(\rho^{(12)}) &= \{f^{(13)}_{03}, f^{(11)}_{03}, \rho_{33}\},
\end{align*}
\]

are found for the set of rotations given by Eq. (43), where the auxiliary function $f_{mn}^{(kl)}$ are defined by

\[
f_{mn}^{(kl)} = \frac{1}{2}(\rho_{mm} + i^n \rho_{nn} + i^k \rho_{nm} + \rho_{mn}).
\]

By using the observation approach I, all these equations can determine the matrices $A'$ (and then $C'$), which are based on the set of $N_{\text{eps}} = 48$ equations given by Eq. (28). The singular values of $C'$ are found to be $\text{svd}(C') = \{12, 8^{0.5}, 7^{0.12}\}$, where our compact notation $\sigma_i^{\text{eps}}$ denotes that $\sigma_i$ occurs $n$ times. Thus, we can determine the condition numbers describing the error robustness of the QST method as $\kappa_2(C') = 6$ and $\kappa_F(C') = 34.24$.

By applying the realistic observation approach II, corresponding to Eqs. (29), we obtain $C''$ having the following singular values:

\[
\text{svd}(C''') = \{48, 24.25, 16.17, 9.97, 6.5, 5.45, 5^{0.5}, 4.91, 4.37, 3^{0.5}, 2.92, 2.26, 2, 1.71\},
\]

which yield the condition numbers $\kappa_2(C''') = 28.14$ and $\kappa_F(C''') = 68.99$.

It is usually recommended to add the identity operation to a given QST method to provide a baseline for noise estimation. Then the observation approach I for the extended set of equations (44) results in the following singular values $\text{svd}(C') = \{13, 9^{0.5}, 2^{0.12}\}$ for which $\kappa_2(C') = 6.5$ and $\kappa_F(C') = 37.41$. The observation approach II yields

\[
\text{svd}(C''') = \{52, 27.59, 18.14, 10.41, 6.5, 4.99, 5^{0.5}, 4.93, 4.42, 3^{0.5}, 2.93, 2.34, 2, 1.74\},
\]

which implies $\kappa_2(C''') = 29.87$ and $\kappa_F(C''') = 74.38$. Thus, one can again conclude that the error robustness of the method, as quantified by the condition numbers $\kappa(C')$ and $\kappa(C'')$, is slightly worse than that for the original method, given by Eq. (43).

VII. CONSTRUCTION OF OPTIMIZED SETS OF OPERATIONS

The error-robustness analysis is based on the properties of the coefficient matrices $C$ (or $A$) and, thus, enables to find experimental setups for the reliable QST even without specific experimental data. The optimization in our approach resides in (i) replacing degenerate multi-photon (multi-quantum) rotations by single-photon ones; and (ii) finding sets of rotations with the minimal number of readouts, which still enable reconstruction of reliable density matrices. Thus, in the following, with the help of the condition numbers $\sigma_{\text{min}}(C)$, $\kappa_2(C)$, and $\kappa_F(C)$, we compare the robustness against errors in measurements of various tomographic methods in the quest to find the optimal sets of tomographic rotations.

A. Single-photon replacements for multi-photon rotations

The discussed sets of rotations for QST include single-photon $X$-rotations ($X_{01}, X_{12}, X_{23}$), two-photon $X$-rotations ($X_{02}, X_{13}$), and a three-photon $X$-rotation ($X_{03}$) together with analogous $Y$-rotations. The methods look very intuitive but they are not the simplest to be realized experimentally due to the degeneracy between $\omega_{03}/3$ and $\omega_{12}$ if the 2nd order quadrupolar shifts are neglected (see Fig. 1). Namely, we want to perform the three-photon rotations $Y_{03}(\pi/2)$ and $X_{03}(\pi/2)$ between levels $|0\rangle$ and $|3\rangle$ (for brevity, we say the 0-3 rotation) solely without changing populations between levels $|1\rangle$ and $|2\rangle$. We can effectively rotate 1-2 without rotating 0-3, but we are not able to rotate 0-3 without rotating 1-2. So a feasible tomographic method should be described without direct rotations 0-3. Under this requirement, it is easy to show analytically that one needs combinations of at least two rotations for some of the operations for
complete reconstruction. Then, unfortunately, the above interpretation of the tomographic operations, given by Eq. (43), loses its clarity.

First, we calculate replacements for multi-photon X-rotations. By inspection, we find that

\[ X_{0n}(\theta) = Y_{ln}(\pi) X_{01}(\theta) Y_{ln}(-\pi) = Y_{01}(\pi) X_{1n}(-\theta) Y_{01}(-\pi) = Y_{02}(\pi) X_{2n}(-\theta) Y_{02}(-\pi) = Y_{2n}(\pi) X_{02}(\theta) Y_{2n}(-\pi) = Y_{01}(\pi) Y_{2n}(\pi) X_{12}(-\theta) Y_{2n}(-\pi) Y_{01}(-\pi). \]  

(46)

Analogously, other replacements can be found. By repeatedly applying the first relation in Eq. (46) we obtain a general formula for any two levels \( k < n \):

\[ X_{kn}(\theta) = Y_{k+1,n}(\pi) X_{k,k+1}(\theta) Y_{k+1,n}(-\pi). \]  

(47)

Alternatively, one can apply the 2nd relation in Eq. (46) to obtain:

\[ X_{kn}(\theta) = Y^{(k,n)} X_{n-1,n}(s\theta) (Y^{(k,n)})^\dagger, \]  

where \( s = (-1)^{n-k+1} \) and

\[ Y^{(k,n)} \equiv Y_{k+1,n}(\pi) Y_{k+1,k+2}(\pi) \cdots Y_{n-2,n}(\pi). \]  

(49)

Note that \( Y_{kn}(\pi) = Y_{k,n}(\pi) \). In the same way, we find replacements for multi-photon Y-rotations for any \( \theta \) and \( k < n \):

\[ Y_{kn}(\theta) = Y_{k+1,n}(\pi) Y_{k+1,k+1}(\theta) Y_{k+1,n}(-\pi) = Y^{(k,n)} Y_{n-1,n}(s\theta) (Y^{(k,n)})^\dagger \]  

(50)

in terms of the pulse sequences given by Eq. (49).

In a special case, for a given QST method of the qudit system, the X-rotations based on three-photon (0-3) and two-photon (0-2 and 1-3) transitions can be replaced by various sequences of rotations requiring only single-photon transitions, e.g.,

\[ X_{03}(\theta) = Y_{01}(\pi) X_{23}(\pi) X_{12}(-\theta) Y_{23}(-\pi) Y_{01}(-\pi), \]
\[ X_{02}(\theta) = Y_{01}(\pi) X_{12}(-\theta) Y_{01}(-\pi), \]
\[ X_{13}(\theta) = Y_{12}(\pi) X_{23}(-\theta) Y_{12}(-\pi), \]  

(51)

and analogously for the multi-quantum Y-rotations.

Finally, we point out some practical aspects in the described realization of a nanometer-scale device in a relation to the problem of degeneracy between \( \omega_{03}/3 \) and \( \omega_{12} \). The rotation frequency is proportional to the 1st Bessel function of the oscillation field strength for the coherent rotation between levels \( |1\rangle \) and \( |2\rangle \), but proportional to the 3rd Bessel function for that between \( |0\rangle \) and \( |3\rangle \). Therefore, the 0-3 rotation becomes negligible if the applied field is weak. Moreover, it is possible to select the oscillating field strength, which satisfies some angle rotation for 0-3, which is different from a multiple of 2\( \pi \) rotation for 2-3. Therefore, it is possible to realize a pure 0-3 operation without 2-3 rotation. However, current amplitude necessary for this operation might be very high and the operation is not realistic from the view point of heating. Another QST method based on sequences of the two-photon pulses \( X_{02}, X_{13}, Y_{02} \) and \( Y_{13} \) would also be experimentally feasible. But usually rotations between the closest levels are much faster and easier to perform.

QST method 2

The simplest theoretical solution to omit rotations requiring three-photon transitions in operations \( R^{(11)} \) and \( R^{(12)} \), given by Eq. (43), is to express them as combinations of three one-photon and two-photon rotations as described above. However, we find that combinations of only two rotations can be used, which additionally improve the error robustness of the QST method. Thus, we suggest the following set of rotations:

\[ R^{(11)} = X_{01}(\frac{\pi}{4}) Y_{13}(\pi), \]
\[ R^{(12)} = Y_{01}(\frac{\pi}{4}) Y_{13}(\pi), \]  

(52)

while the other \( R^{(k)} (k = 1, ..., 10) \) are the same as in the QST method 1. It is seen that the new operations do not require the rotation 0-3. In the observation approach I, the singular values \( \text{svd}(C') \), and thus the condition numbers, are the same as in the QST method 1. However, the observation approach II leads to another set of singular values:

\[ \text{svd}(C'') = \{48, 23.48, 14.25, 9.79, 6.5, 5.45, 5^\otimes 3, 4.996, 4.70, 4.22, 3^\otimes 2, 2.90, 2.21\}. \]

Note that \( \sigma_{\text{max}}(C'') \) is the same in both methods, while \( \sigma_{\text{min}}(C'') \) is greater than that in the QST method 1. Thus, the condition numbers \( \kappa_2(C'') = 21.67 \) [and also \( \kappa_F(C'') = 54.80 \)] are closer to 1 implying that the robustness against errors of the new method is improved.

A modified method can be obtained from the method based on Eq. (52) on inclusion of the identity operation, \( R^{(0)} = I \). Then the observation approach I leads to the same error robustness as that for the analogous modified QST method 1. In contrast to this approach, the observation approach II yields: \( \sigma_{\text{max}}(C'') = 52, \sigma_{\text{min}}(C'') = 2.30 \) and \( \kappa_2(C'') = 22.64 (\kappa_F(C'') = 58.998) \). Thus, we conclude (see also Table I) that the \( \sigma_{\text{max}}(C'') \) and \( \sigma_{\text{min}}(C'') \) are increased in comparison to those obtained from the method given by Eq. (52), but in such a way that the error robustness of the QST, as quantified by the condition numbers, is slightly higher.

We mention that the two-photon transitions used in rotations given by Eq. (52) can also be replaced by single-photon transitions with the help of the sequences of rotations given by Eq. (51).
By recalling Eq. (45), one can easily find that the diagonal elements of the matrices $A$ and $C$, obtained from Eq. (54), determine the error robustness of the QST method depending on the observation approach. In the observation approach I, the singular values are highly degenerate, as $\text{svd}(C') = \{6, 2^{5/2}\}$, yielding $\kappa_2(C') = 3$ and $\kappa_F(C') = 19.04$. In the observation approach II, one obtains: $\text{svd}(C'') = \{24, 6.83^{8/5}, 6.4^{5/2}, 2.1.17^{15/7}\}$, and so the condition numbers are $\kappa_2(C'') = 20.49$ and $\kappa_F(C'') = 56.48$. In comparison to the former methods, we conclude that the error robustness, as measured by the condition numbers, is much better (even twice better for $\kappa_2$).

One can consider various modifications of the method, e.g., by choosing other combinations of $X$ and $Y$ rotations to mention:

$$R^{(1)} = X_0 Y_{22}, \quad R^{(4)} = Y_0 X_{22},$$
$$R^{(2)} = X_0 Y_{12}, \quad R^{(5)} = Y_0 X_{12},$$
$$R^{(3)} = X_0 Y_{13}, \quad R^{(6)} = Y_0 X_{13},$$

(55)

for which the error robustness of the method is unchanged.

Analogously to our modifications of the QST method I, we include the identity operation, $R^{(0)} = \mathbb{I}$, in the set of rotations in the preceding method. As a result, we obtain less degenerate singular values of $C$. Specifically, by applying the observation approach I, one obtains: $\text{svd}(C') = \{7, 3^{3/4}, 2^{9/2}\}$, which yields $\kappa_2(C') = 3.5$ and $\kappa_F(C') = 20.39$. For the observation approach II we obtain: $\text{svd}(C'') = \{28, 9.70, 6.93, 6.83^{8/5}, 6^{5/2}, 4^{3/4}, 2^{\frac{5}{2}}, 1.37, 1.17^{15/7}\}$, which gives $\kappa_2(C'') = 23.90$ and $\kappa_F(C'') = 57.92$. In conclusion, $\sigma_{\min}(C) (C = C', C'')$ is the same for the original method 3 and its modification, but $\sigma_{\max}(C)$ is increased. Thus, the error robustness is slightly worse in comparison to the QST method 3, independent of the observation approach.

### B. QST with the minimal number readouts

#### QST method 3

An analysis of the rotations $R^{(k)}$, given by Eq. (43) or, equivalently, of the transformed diagonal elements $\text{diag}(\rho^{(k)})$, given by Eq. (43), shows that in each rotation only two elements are modified. To make QST more efficient, we can apply such rotations to modify all four diagonal elements. Thus, e.g., one can combine the following rotations (as shown in Fig. 4):

$$R^{(1)} = Y_0 Y_{23}, \quad R^{(2)} = X_0 X_{23},$$
$$R^{(3)} = Y_0 Y_{13}, \quad R^{(4)} = X_0 X_{13},$$
$$R^{(5)} = Y_0 Y_{12}, \quad R^{(6)} = X_0 X_{12}.$$  

(53)

By recalling Eq. (45), one can easily find that the diagonal elements of $\rho$ after the $k$th rotation, given by Eq. (53), are described by:

$$\text{diag}(\rho^{(1)}) = [f^{(22)}_{01}, f^{(00)}_{01}, f^{(22)}_{23}, f^{(00)}_{23}],$$
$$\text{diag}(\rho^{(2)}) = [f^{(13)}_{01}, f^{(31)}_{01}, f^{(13)}_{23}, f^{(31)}_{23}],$$
$$\text{diag}(\rho^{(3)}) = [f^{(22)}_{02}, f^{(22)}_{02}, f^{(00)}_{02}, f^{(00)}_{02}],$$
$$\text{diag}(\rho^{(4)}) = [f^{(23)}_{02}, f^{(13)}_{02}, f^{(31)}_{02}, f^{(31)}_{02}],$$
$$\text{diag}(\rho^{(5)}) = [f^{(22)}_{03}, f^{(22)}_{03}, f^{(00)}_{03}, f^{(00)}_{03}],$$
$$\text{diag}(\rho^{(6)}) = [f^{(13)}_{03}, f^{(13)}_{03}, f^{(31)}_{03}, f^{(31)}_{03}],$$

(54)

where $f^{(k)}_{mn}$ is defined by Eq. (45). The coefficient matrices $A$ and $C$, obtained from Eq. (54), determine the error robustness of the QST method depending on the observation approach. In the observation approach I, the singular values are highly degenerate, as $\text{svd}(C') = \{6, 2^{5/2}\}$, yielding $\kappa_2(C') = 3$ and $\kappa_F(C') = 19.04$. In the observation approach II, one obtains: $\text{svd}(C'') = \{24, 6.83^{8/5}, 6.4^{5/2}, 2.1.17^{15/7}\}$, and so the condition numbers are $\kappa_2(C'') = 20.49$ and $\kappa_F(C'') = 56.48$. In comparison to the former methods, we conclude that the error robustness, as measured by the condition numbers, is much better (even twice better for $\kappa_2$).

One can consider various modifications of the method, e.g., by choosing other combinations of $X$ and $Y$ rotations to mention:

$$R^{(1)} = X_0 Y_{23}, \quad R^{(4)} = Y_0 X_{23},$$
$$R^{(2)} = X_0 Y_{13}, \quad R^{(5)} = Y_0 X_{13},$$
$$R^{(3)} = X_0 Y_{12}, \quad R^{(6)} = Y_0 X_{12},$$

(55)

for which the error robustness of the method is unchanged.

Analogously to our modifications of the QST method I, we include the identity operation, $R^{(0)} = \mathbb{I}$, in the set of rotations in the preceding method. As a result, we obtain less degenerate singular values of $C$. Specifically, by applying the observation approach I, one obtains: $\text{svd}(C') = \{7, 3^{3/4}, 2^{9/2}\}$, which yields $\kappa_2(C') = 3.5$ and $\kappa_F(C') = 20.39$. For the observation approach II we obtain: $\text{svd}(C'') = \{28, 9.70, 6.93, 6.83^{8/5}, 6^{5/2}, 4^{3/4}, 2^{\frac{5}{2}}, 1.37, 1.17^{15/7}\}$, which gives $\kappa_2(C'') = 23.90$ and $\kappa_F(C'') = 57.92$. In conclusion, $\sigma_{\min}(C) (C = C', C'')$ is the same for the original method 3 and its modification, but $\sigma_{\max}(C)$ is increased. Thus, the error robustness is slightly worse in comparison to the QST method 3, independent of the observation approach.

#### QST method 4

To avoid three-photon transitions, required for method 2, one can apply our replacements based on single- and two-photon transitions, given by Eq. (41). One can even further simplify such methods to obtain, e.g.,

$$R^{(3)} = X_0 Y_{13}^2 X_{12},$$
$$R^{(6)} = Y_0 Y_{13} Y_{12},$$

(56)

where $Y_{13}^2 \equiv Y_{13}(\pi)$ and the other 4 rotations are the same as in the QST method 3. In the observation approach I, the singular values $\text{svd}(C')$ are found to be the same as in the QST method 3, however the observation approach II predicts not completely the same singular values as $\text{svd}(C'') = \{24, 6.83^{8/5}, 6^{5/2}, 4^{3/4}, 2^{\frac{5}{2}}, 1.37, 1.17^{15/7}\}$. It is interesting to observe that $\sigma_{\min}(C'')$ and $\sigma_{\max}(C'')$ are the same for both methods, some other values are different resulting in $\kappa_2(C'') = 20.49$ and $\kappa_F(C'') = 51.14$. Thus, the error robustness measured by $\kappa_2$ is unchanged, although that described by $\kappa_F(C'')$ is slightly improved in comparison to the QST method 3.
On including the identity operation, $R(0) = I$, to the rotations of the method, we obtain the same svd$(C^\prime)$ for the observation approach I. However, the observation approach II yields some singular values different from those in the QST method 4, i.e., svd$(C^\prime\prime) = \{28, 9.70, 7.24, 6.93, 6^{\otimes 2}, 4^{\otimes 6}, 2.76, 2^{\otimes 2}, 1.37\}$. Note that $\sigma_{\text{min}}(C^\prime\prime)$ is increased, while $\sigma_{\text{max}}(C^\prime\prime)$ is kept unchanged. Thus, we obtain $\kappa_2(C^\prime\prime) = 20.47$ and $\kappa_F(C^\prime\prime) = 43.79$, which are lower than those parameters in the QST method 4. It is seen (as summarized in Table I) that the error robustness of this method is slightly improved in comparison to all former methods for the observation approach II.

As discussed for the QST method 2, the corresponding sets of rotations for the present method can include solely single-photon transitions if one applies the substitution rules given by Eq. (51).

### VIII. GENERALIZED QST FOR HIGHER-SPIN SYSTEMS

It is highly desirable to scale quantum information processing beyond two logical qubits (spin-3/2 system) and, thus, to develop QST for such systems.

Our former $M_2$-based QST methods described for spin-3/2 nuclei can readily be generalized to quadrupolar nuclei with an arbitrary spin number $I$, which have $N = 2I + 1$ non-equidistant energy levels in an external magnetic field due to the Zeeman and quadrupolar interactions.

A formally simple $M_2$-based QST method of a density matrix of the $N$-level system can be composed of $N(N - 1)$ readouts of the following rotations: $(N - 1)$ single-photon rotations $(X_{01}, X_{12}, ..., X_{N-1,N})$, $(N - 2)$ two-photon rotations $(X_{02}, X_{13}, ..., X_{N-2,N})$, etc., and a $(N - 1)$-photon rotation $X_{0N}$, together with analogous $Y$-rotations. Note that the multi-photon rotations can be replaced by single-photon rotations according to the substitution rules, given by Eqs. (46)–(51).

#### A. QST of spin-7/2 systems

In a special case of the general approach to the $M_2$-based QST, let us consider QST methods for a spin-7/2 system, which in an external magnetic field $B_0$ has eight energy levels, and referred to as the eight-dimensional qudit or quocit, as discussed in Sec. III.

A few theoretical proposals and NMR experiments were performed on liquid-state spin-7/2 systems: a theoretical proposal of CCNOT gate [61], a preparation of effective pure states [35], quantum simulation [36], half-adder and subtractor operations [27, 47], test of phase coherence in electromagnetically-induced transparency [38]. One can easily list a few other quantum algorithms, which could be realized in spin-7/2 NMR experiments offering much more refined tests of quantum-information principles in comparison to such realizations on spin-3/2 nuclei. As discussed in subsection III.C, some gates (e.g., the Toffoli gate) in an eight-level system can be implemented in a much simpler way than those in a real three-qubit system.

An $M_{xy}$-based QST for a spin-7/2 system was applied by Teles et al. [20]. Here, we describe QST methods of spin-7/2 systems based on the $M_x$-detection.

#### QST method 5

The QST of $\rho$ of an eight-level system can be based on 56 readouts of the following natural set of rotations:

$$(R(k))_k = \{(Y_{01}, Y_{12}, Y_{34}, Y_{45}, Y_{56}, Y_{67}), (Y_{02}, Y_{13}, Y_{24}, Y_{35}, Y_{46}, Y_{57}), (Y_{03}, Y_{14}, Y_{25}, Y_{36}, Y_{47}), (Y_{04}, Y_{15}, Y_{26}, Y_{37}), (Y_{05}, Y_{16}, Y_{27}), (Y_{06}, Y_{17}), (Y_{07})\}, \quad (57)$$

and analogously for $X$-rotations. Brackets are added to indicate groups of $n$-photon ($n = 1, ..., 7$) rotations. As in the former methods for a given observation approach, one can determine matrices $A$ and then $C$ together with their condition numbers. To avoid rotations requiring multi-photon transitions, one can express them solely by single-photon ones according to the replacement rules given by Eqs. (48) and (50). Note that the condition numbers are unchanged by this procedure.

According to the observation approach I based on the set of $N_{\text{eq}} = 8 \times 56 = 448$ equations given by [28], we find all the 64 singular values $\sigma_i(C^\prime)$ to be highly degenerate, i.e., svd$(C^\prime) \equiv \{\sigma_i(C^\prime)\} = \{56, 48^{\otimes 7}, 2^{\otimes 56}\}$. The corresponding condition number based on the spectral norm is $\kappa_2(C^\prime) = 28$, while that based on the Frobenius norm is found to be $\kappa_F(C^\prime) = 552.4$. According to the observation approach II based on 448 equations given by [29], we find that the 64 singular values $\sigma_i(C^\prime\prime)$ are much less degenerate than $\sigma_i(C^\prime)$ and they range from $\sigma_{\text{min}}(C^\prime\prime) = 1.27$ to $\sigma_{\text{max}}(C^\prime\prime) = 448$. The resulting condition numbers are $\kappa_2(C^\prime\prime) = 353.96$ and $\kappa_F(C^\prime\prime) = 1196.97$.

In comparison to the analogous QST method I for QST of a four-level system, it is seen that the present method requires determination of 64 real unknowns (instead of 16) via 56 readouts (instead of 12 in the former method). This results in much worse robustness to errors in the measured data for the present method, e.g., the condition numbers $\kappa_2(C^\prime\prime)$ and $\kappa_F(C^\prime\prime)$ are greater by three-orders than those in the former method for a quocit.

#### QST method 6

By combining properly the above rotations, we propose an optimized tomographic method based only on 14 readouts (as shown in Fig. 5):

$$R^{(1)} = Y_{01} Y_{23} Y_{45} Y_{67}.$$
and the other $R^{(k)}$ for $k = 8, \ldots, 14$ are defined analogously using $X$-rotations.

The observation approach I leads to the following set of highly degenerate singular values $\text{svd}(C') \equiv \{\sigma_i(C')\} = \{14, 6^{\otimes 7}, 2^{\otimes 56}\}$ and the corresponding condition numbers: $\kappa_2(C') = 7$ (which is $< 28$ obtained for the not-optimized method) and $\kappa_F(C') = 97.68$ ($< 522.4$). The observation approach II results in less degenerate singular values with the extremal ones given by $\sigma_{\min}(C'') = 0.384$ to $\sigma_{\max}(C'') = 112$. So, the condition numbers are $\kappa_2(C'') = 291.48$ ($< 353.96$) and $\kappa_F(C'') = 636.1$ ($< 1191.97$). Thus, we find (see also Table 1) that by applying our optimization method, the number of readouts is decreased and the robustness against errors of the methods is improved, as measured by the condition numbers based on the spectral and Frobenius norms, becomes lower.

It should be noted again that the rotations in Eqs. (22) based on multi-photon transitions can be expressed by combinations of single-photon rotations according to Eqs. (48) and (50). The condition numbers are the same in such modifed methods. Further simplifications can be obtained by omitting some of the single-photon rotations, which can also result in better robustness to errors.

FIG. 5: Tomography of an eight-level system with the optimized number of readouts, as given by Eq. (58). There are altogether 14 readouts including the seven $X$-rotations and the corresponding seven $Y$-rotations (shown by arrows). Note that the standard method, given by Eq. (57), requires 56 readouts. Pulses in boxes can be applied simultaneously or sequentially.

\[
R^{(2)} = Y_{02}Y_{13}Y_{46}Y_{57}, \\
R^{(3)} = Y_{03}Y_{16}Y_{25}Y_{47}, \\
R^{(4)} = Y_{04}Y_{15}Y_{26}Y_{37}, \\
R^{(5)} = Y_{05}Y_{14}Y_{27}Y_{36}, \\
R^{(6)} = Y_{06}Y_{17}Y_{24}Y_{35}, \\
R^{(7)} = Y_{07}Y_{12}Y_{24}Y_{56},
\]

FIG. 6: (Color online) An example of an unphysical density matrix $\rho$ reconstructed by a linear tomography from the simulated experimental data using the QST method 4 (or, equivalently, the QST method 1): (a) real and (b) imaginary parts of the elements $\rho_{mn}$, corresponding to Eq. (59).

**IX. NUMERICAL SIMULATIONS AND MAXIMUM-LIKELIHOOD METHOD**

Here, we present a numerical simulation of the experimental QST based on the $M_z$-detection for a quartit.

Let us assume that by applying the QST method 4, given by Eq. (50), with the observation approach II one has obtained the following set of experimental data $\langle b^{(k)}_{i,k} \rangle_{n,k} = (0.0228, -0.0166, 0.0268, -0.0037, -0.0320, 0.0168, -0.0760, -0.4658, 0.0292, -0.0811, -0.0484, -1.0384, 0.0570, -0.0542)$. By applying the linear reconstruction described in Sec. V, we obtained an unphysical density matrix $\rho$, given by Eq. (62) and shown in Figs. 4(a,b). It is unphysical as it has negative eigenvalues (i.e., equal to -0.065 and -0.024). Note that we chose data $b^{(k)}_{i,k}$ in such a way that Eq. (C2) corresponds exactly to $\rho$ given by Eq. (3.21) in Ref. [4].

A linear reconstruction analysis applied to experimental data often results in the prediction of a unphysical density matrix, which can be a non-positive operator (as shown in Fig. 6). We note that a linear QST method, even if it is optimally robust to errors (as quantified by condition numbers equal to 1), can sometimes lead to unphysical density matrices. Thus, we show how to apply a nonlinear reconstruction method for our NMR QST methods to overcome this problem. Our approach is based on the maximum-likelihood (MaxLik) method for a QST method, described by, e.g., James et al. [4], of two polarization degrees of entangled photons. For simplicity, we are not using the more efficient MaxLik algorithms of, e.g., Ref. [62]. Here, for brevity, we focus on a QST of a quartit system. The generalization for spin-$(2n + 1)/2$ system is straightforward.

A “physical” (i.e., normalized, Hermitian, and positive) density matrix $\rho_{\text{phys}}$ can be constructed explicitly to be of the form

\[
\rho_{\text{phys}}(T) = \frac{T^{\dagger}T}{\text{tr}(T^{\dagger}T)},
\]

where $T$ is the detector Mueller matrix.
where $T$ is the $4 \times 4$ lower-triangular (or, equivalently, upper-triangular) matrix, which is easily invertible. The diagonal terms are assumed to be real, while the off-diagonal terms are chosen to be complex. By the Schur decomposition, any normal matrix $F$ (i.e., $F^\dagger F = F F^\dagger$) can be represented as $F = U T U^\dagger$ in terms of a triangular matrix $T$ and a unitary matrix $U$. For simplicity, we neglect $U$.

Let us assume a Gaussian noise for the measurements. Thus, a likelihood function can be chosen to be the normal distribution

$$P(b, T) = N^T \prod_{k=1}^{N_r} \prod_{n=1}^{N_{vals}} \exp \left( \frac{[b^{(k)}_n] - \tilde{b}^{(k)}_n(T))^2}{2\sigma^{(k)}_n(T)^2} \right)$$

(60)

describing the probability of the measured elements of the observation vector $b \equiv [b^{(k)}_n]_{n,k}$ for a given $\rho_{\text{phys}}(T)$, where $N_{\text{vals}} = 4$ and $\tilde{b}^{(k)}_n(T) = [\rho^{(k)}_{\text{phys}}(T)]_{n,n}$ for the observation approach I, while for the observation approach II we have $N_{\text{vals}} = 3$ and

$$\tilde{b}^{(k)}_n(T) = \text{Tr}[I^{(n+1,n)}_Z \rho^{(k)}_{\text{phys}}(T)],$$

$$I^{(n+1,n)}_Z = [n + 1](n + 1) - [n][n].$$

(61)

Moreover, $\rho^{(k)}_{\text{phys}}(T) = R(k)_{\text{phys}}(R(k)^{\dagger})_{\text{phys}}$, $\sigma^{(k)}_n(T)$ is the corresponding standard deviation, and $N^T$ is the normalization constant, assumed to be independent of $T$. As usual, $N_r$ is the number of readouts assuming that each gives $N_{\text{vals}}$ values.

An optimized physical density matrix $\rho_{\text{phys}}(T)$ can be found by numerically searching the maximum $\max_T P(b, T)$ for a given observation vector $b$ or, equivalently, $\max_T \log P(b, T)$, which corresponds to finding a minimum

$$\mathcal{L}_{\text{min}} = \min_T \mathcal{L}(b, T)$$

(62)

of the following likelihood function:

$$\mathcal{L}(b, T) = \sum_{k=1}^{N_r} \sum_{n=1}^{N_{\text{vals}}} \left( \frac{b^{(k)}_n - \tilde{b}^{(k)}_n(T)}{\sigma^{(k)}_n(T)} \right)^2.$$  

(63)

The initial matrix $T_{\text{ini}}$, required for the numerical optimization, can be estimated from our “unphysical” $\rho$ (reconstructed with the standard linear method) by inverting Eq. (59), which results in [3]:

$$T_{\text{ini}} = \begin{bmatrix}
N_1 d & 0 & 0 & 0 \\
N_2 d'_{01} & N_2 d'_{00} & 0 & 0 \\
N_3 d'_{01,12} & N_3 d'_{00,12} & N_3 d'_{00,11} & 0 \\
N_4 \rho_{33,0} & N_4 \rho_{31,0} & N_4 \rho_{31,0} & N_4 \rho_{32} & N_4 \rho_{33}
\end{bmatrix},$$

(64)

where $d = \det(\rho)$, $d'_{ij} = (d'_{ij})_{1,2,3,4}$ is the first (second) minor of $\rho$, and the normalization factors are

$$N_1 = \frac{1}{\sqrt{d'_{00,0}d}}, \quad N_2 = \frac{1}{\sqrt{d'_{00,0,11}d'_{00,0,11}}}, \quad N_3 = \frac{1}{\sqrt{\rho_{33}d'_{00,0,11}}},$$

(65)

FIG. 7: (Color online) The real parts of the physical density matrices reconstructed by the maximum-likelihood method from the simulated experimental data: (a) $\rho^\prime_{\text{phys}, v} \equiv \rho^\prime_{\text{phys}, v} \equiv \rho^\prime_{\text{phys}, v}$, corresponding to Eq. (C3), obtained using the QST method 4, and (b) $\rho^\prime_{\text{phys}, \text{un}} \equiv \rho^\prime_{\text{phys}, \text{un}} \equiv \rho^\prime_{\text{phys}, \text{un}}$, corresponding to Eq. (C4), based on the QST method 1. The imaginary parts of $\rho'$ and $\rho''$ are close to zero [cf. Fig. 6(b)] and thus, for brevity, are not presented.

Our numerical minimization of the likelihood function $\mathcal{L}_{\text{min}}$, for the simulated experimental data of this section, resulted in the values listed in Table II of Appendix C. In particular, the numerical minimalization assuming the method 4 enabled us to decrease the likelihood function $\mathcal{L}(b, T)$ from 3.43 to 0.16, and the corresponding optimized physical density matrix $\rho_{\text{phys}}^\prime$ is given by Eq. (C3). The real part of $\rho_{\text{phys}}^\prime$ is presented in Fig. 7(a), and is compared with the unphysical matrix $\rho$, given by Eq. (C2), in Fig. 8(a).

As another example, let us assume that by applying the QST method 1, given by Eq. (C3), we obtained data (for brevity not presented here), which after the linear reconstruction again lead to an unphysical density matrix $\rho$, given by Eq. (C2). However, by applying our numerical minimalization procedure within the maximum-likelihood method, we obtain the physical density matrix $\rho_{\text{phys}}^\prime$ given by Eq. (C4). As expected, the matrix $\rho_{\text{phys}}^\prime$ slightly differs from $\rho_{\text{phys}}$. In this example, the likelihood function $\mathcal{L}(b, T)$ was diminished from 11.09 to 0.15 (as shown in Table II). Analogously to the $\text{Re}(\rho_{\text{phys}}^\prime)$ presented in Figs. 5(a) and 6(a), $\text{Re}(\rho_{\text{phys}})$ is shown in Fig. 7(b) and compared with the unphysical matrix $\rho$ in Fig. 8(b).

X. CONCLUSIONS

We described various methods for implementing quantum state tomography for systems of quadrupolar nuclei with spins 3/2 (equivalent to quartz) and 7/2 (quoctit) in an unconventional approach to NMR, which is based on the measurement of longitudinal magnetization $M_z$ instead of the standard measurement of the transverse magnetization $M_x$. This work has been motivated by the demonstration of high-precision $M_z$-based NMR techniques of coher-
ent manipulation of nuclear spins $J = 3/2$ ($^{69}$Ga, $^{71}$Ga, and $^{75}$As) in a GaAs quantum-well device based on an the fractional quantum Hall effect [42]. The device, exhibiting extremely low decoherence [44], offers new possibilities to study interactions in semiconductors but also enables the realization of single- and two-qubit quantum gates [43] and, possibly, testing simple quantum-information processing algorithms.

Although our presentation of the protocols of QST of large-nuclear systems was focused on the nanoscale semiconductor device of Ref. [42], it should be stressed that these protocols can also be readily applied to large-nuclear spins in other systems.

We proposed methods with optimized sets of rotations. The optimization was applied in order to decrease the number of NMR readouts and to improve the robustness against errors, as quantified by condition numbers.

Some of the proposed QST methods for a quartit system require the three-photon transitions (between levels $|0\rangle$ and $|3\rangle$), which are induced by relatively strong pulses. Unfortunately, such pulses can simultaneously induce transitions between levels $|1\rangle$ and $|2\rangle$. Thus, from a practical point of view, it is desired to apply only weak pulses selectively inducing single-photon transitions. We showed how the rotations requiring multi-photon transitions can be replaced by combinations of rotations based only on single-photon transitions.

By applying matrix-condition numbers [50, we compared robustness against errors in the measured data of all the described tomographic methods. Table I presents the condition numbers $\kappa_2$ and $\kappa_F$ for all the discussed QST methods for systems with both spins-3/2 and spins-7/2. We have assumed three observation approaches corresponding to: (I) an ideal $M_z$-detection, where all diagonal elements $\rho_{nn}$ ($n = 0, \ldots, 3$) of a density matrix can be directly accessed; (II) a realistic $M_z$-detection, where the population differences ($\rho_{11} - \rho_{00}$, $\rho_{22} - \rho_{11}$, and $\rho_{33} - \rho_{22}$) can be estimated from the amplitude of the signals by integrating the area of the peaks centered at $\omega_{01}$, $\omega_{12}$, and $\omega_{23}$ (see Fig. 2), respectively; and (III) the CYCLOPS method, where the information gathered from $M_z$-spectra corresponds to some linear functions of the diagonal elements $\rho_{nn}$, as given by Eq. (32). Table I shows that the more applied is the method the worse is the error robustness, as measured by the condition numbers. The ideal observation approach I is very robust to errors. In contrast to this, the condition numbers for the observation approach III are so large, so one can conclude that the corresponding QST methods are ill-conditioned. Fortunately, the QST methods based on the realistic observation approach II are relatively well-conditioned.

By analyzing Table I, one can conclude that the tomographic methods 3 and 4, which are based on smaller number of readouts, also exhibit much improved robustness against errors. Indeed these methods are described by the condition numbers $\kappa_2$, which are approximately equal to 1/2, 3/4, and 2/3 of $\kappa_2$ using the methods 1 and 2 for the observation approaches I, II, and II, respectively.

Let us now compare the error robustness in terms of $\kappa_2$ for the discussed NMR QST methods for a quartit (i.e., two virtual qubits) with some known optical QST methods (see, e.g., Ref. [8] for a review) for two physically-distinct qubits: The well-known QST protocol of James et al. [4], which is solely based on local measurements, yields the condition number $\kappa_2 = 60.1$. The QST of Refs. [5, 6] is based on the standard separable basis composed of the 36 two-qubit eigenstates of the tensor products of the Pauli operators. This often-applied QST yields $\kappa_2 = 9$. In contrast to this, the recently-proposed QST of Ref. [8], which is based on local and global measurements of generalized Pauli operators, is optimal since it yields the condition number $\kappa_2 = 1$. Another QST, which is based on local measurements of the 16 tensor products of the Pauli operators, yields $\kappa_2 = 2$.

For the QST methods using the observation approach II (see Table I), the condition numbers $\kappa_2$ range from approx. 20 (for the methods 3 and 4) to 30 (for the methods 1 and 2). This means that these methods are twice or even three times more robust to errors than the QST method of James et al. [4]. The error-robustness of the QST of Refs. [5, 6] is better than that for our methods based on the observation approach II. However, if the observation approach I is applied, all these NMR methods are superior to these two optical QST methods.

As linear reconstruction can lead to unphysical density matrices for experimental data, we also applied nonlinear reconstruction based on the maximum-likelihood method. Our method is refined in comparison to the standard approach (see, e.g., Ref. [4]), as we performed the minimalization of a likelihood function over a larger class of physical density matrices.

We also described sequences of NMR pulses to perform various quantum tomography methods and arbitrary gates (including single virtual qubit rotations) with nuclear spins of an arbitrary value (equivalent to qudits). This enables a simple translation of arbitrary quantum algorithms from systems of spins-1/2 to higher-number spins.

In addition, we presented an exemplary state reconstruction of a density matrix of a spin-3/2 system from

![FIG. 8: (Color online) Comparison of the unphysical (with elements $\rho_{nn}$) and physical ($\rho'_{nn}$ and $\rho''_{nn}$) density matrices shown in Fig. 7: (a) $\Delta'_{mn} \equiv |\text{Re}(\rho'_{mn} - \rho_{mn})|$ and (b) $\Delta''_{mn} \equiv |\text{Re}(\rho''_{mn} - \rho_{mn})|$.](image-url)
numerically simulated experimental data.

Finally, we express our hope that this comparative study of various NMR tomographic methods will draw attention to the issue of how such methods are robust against errors and, thus, to the question about the reliability of the reconstructed density matrices.

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Appendix A: Selective rotations

Selective rotations in quadrupolar nuclei with large spins are a simple generalization of the standard rotations in a spin-1/2 system:

\[
X(\theta) = R_x(\theta) = \begin{bmatrix} \cos \frac{\theta}{2} & -i \sin \frac{\theta}{2} \\ i \sin \frac{\theta}{2} & \cos \frac{\theta}{2} \end{bmatrix}, \quad Y(\theta) = R_y(\theta) = \begin{bmatrix} \cos \frac{\theta}{2} & \sin \frac{\theta}{2} \\ \sin \frac{\theta}{2} & \cos \frac{\theta}{2} \end{bmatrix}, \quad Z(\theta) = R_z(\theta) = \begin{bmatrix} e^{-i\theta/2} & 0 \\ 0 & e^{i\theta/2} \end{bmatrix}.
\]

If a two-level rotation is \( R^{(i)}(\theta) = \begin{bmatrix} a & b \\ c & d \end{bmatrix} \) (with \( i = X,Y,Z \)), then the corresponding selective rotation between levels \( m < n \) in an \( N \)-level system is given by

\[
R^{(i)}_{mn}(\theta) = a|m\rangle\langle m| + b|m\rangle\langle n| + c|n\rangle\langle m| + d|n\rangle\langle n| + \sum_{k \neq m,n} |k\rangle\langle k|.
\]

For example, the matrix representation of the rotation \( X_{02}(\frac{\pi}{2}) \) in a spin-3/2 system reads as:

\[
X_{02}(\frac{\pi}{2}) = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & -i & 0 \\ 0 & \sqrt{2} & 0 & 0 \\ -i & 0 & 1 & 0 \\ 0 & 0 & 0 & \sqrt{2} \end{pmatrix}.
\]

Note that the rotations calculated by \( \exp(-i\mathcal{H}_{\text{rot}}t_p/\hbar) \) are, in general, not exactly corresponding to Eq. (A4), because these depend on the quadrupolar frequency \( \omega_Q \), even if the conditions \( \hbar \omega^{(k)}_Q = \epsilon_m - \epsilon_n \) and \( |\omega_k| \ll |\omega_Q| \) are satisfied [48]. Nevertheless, these rotations can be effectively reduced to Eq. (A4) if the pulse duration \( t_p \) is equal to \( 2\pi/\omega_Q \) or its multiple. To fulfill this condition experimentally, the line intensities of spectra can be monitored as a function of the pulse duration (see, e.g., Ref. [10]).

Appendix B: \( M_{x_y} \)-detection vs \( M_z \)-detection

The \( M_{x_y} \)-detection of a two spin-1/2 system enables determination of the off-diagonal elements marked in boxes:

\[
\rho = \begin{pmatrix} \rho_{00} & \rho_{01} & \rho_{02} & \rho_{03} \\ \rho_{10} & \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{20} & \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{30} & \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix}, \quad (B1)
\]

On the other hand, the \( M_{xy} \)-detection of a spin-3/2 system provides the other off-diagonal elements (also marked in boxes):

\[
\rho = \begin{pmatrix} \rho_{00} & \rho_{01} & \rho_{02} & \rho_{03} \\ \rho_{10} & \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{20} & \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{30} & \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix}.
\]

In contrast to this, the \( M_z \)-detection of a spin-3/2 system and two spin-1/2 systems enables the determination of only the diagonal elements \( \rho_{ii} \) (\( i = 0, ..., 3 \)). This is because the NMR signals obtained by the \( M_z \)-detection of a spin-3/2 system are given by \( M_z \propto |\text{Tr}[\rho I_z]| \), while those obtained by the \( M_{xy} \)-detection are proportional to [63]:

\[
M_{x_y}^\pm = M_x \pm i M_y \propto \text{Tr}[\rho I_x] \quad (B3)
\]

given in terms of the total angular momentum operator \( I_\pm = I_x \pm i I_y \) for spin \( I = 3/2 \), where

\[
I_x = \begin{pmatrix} 0 & a & 0 & 0 \\ a & 0 & 1 & 0 \\ 0 & 1 & 0 & a \\ 0 & 0 & a & 0 \end{pmatrix}, \quad I_y = i \begin{pmatrix} 0 & -a & 0 & 0 \\ a & 0 & -1 & 0 \\ 0 & 1 & 0 & -a \\ 0 & 0 & a & 0 \end{pmatrix}, \quad (B4)
\]

with \( a = \sqrt{3}/2 \).

Appendix C: Examples of reconstructed density matrices

Here, we present reconstructed density matrices in linear and nonlinear approaches for our numerical experiment of Sec. IX.
TABLE II: Minimized likelihood function $L_{\min}$ for nonlinear reconstructions of states of nuclear spin $I = 3/2$. Key: $L_{\min}$ is given by Eq. (62) for the experimental data simulated in such a way that by application of linear reconstructions lead to the same unphysical density matrix, given by Eq. (C2) for any observation approach (OA). Thus, $L_{\min}$ shows a “distance” between unphysical density matrix and the closest physical density matrix. Other symbols are the same as in Table I.

| method rotations OA | $E_{\min}$ |
|---------------------|-------------|
| method 1 Eq. (43)   | I 0.4162    |
|                     | I* 0.4216   |
|                     | II 0.1488   |
|                     | II* 0.1531  |
| method 2 Eq. (52)   | I 0.4167    |
|                     | I* 0.4214   |
|                     | II 0.2126   |
|                     | II* 0.2181  |
| method 3 Eq. (53)   | I 0.3504    |
|                     | I* 0.3673   |
|                     | II 0.0693   |
|                     | II* 0.0759  |
| method 4 Eq. (56)   | I 0.3504    |
|                     | I* 0.3673   |
|                     | II 0.1635   |
|                     | II* 0.1748  |

Instead of writing $\rho$ explicitly we use the equivalent but more compact matrix $f(\rho)$ defined for a quartit as follows

$$f(\rho) = \begin{bmatrix}
\rho_{00} & \text{Re}\rho_{01} & \text{Re}\rho_{02} & \text{Re}\rho_{03} \\
\text{Im}\rho_{01} & \rho_{11} & \text{Re}\rho_{12} & \text{Re}\rho_{13} \\
\text{Im}\rho_{02} & \text{Im}\rho_{12} & \rho_{22} & \text{Re}\rho_{23} \\
\text{Im}\rho_{03} & \text{Im}\rho_{13} & \text{Im}\rho_{23} & \rho_{33}
\end{bmatrix}.$$  \hspace{1cm} (C1)

The linear reconstruction based on data obtained using the QST methods 1 and 4 results in the following unphysical density matrix:

$$f(\rho) = \begin{bmatrix}
0.4872 & -0.0042 & -0.0098 & 0.5192 \\
-0.0114 & 0.0045 & 0.0271 & -0.0648 \\
0.0178 & 0.0146 & 0.0062 & -0.0695 \\
-0.0380 & 0.0076 & -0.0134 & 0.5020
\end{bmatrix}, (C2)$$

which is shown in Fig. 6. The physical density matrices obtained via the nonlinear reconstruction are found to be:

$$f(\rho')_{\text{phys}} = \begin{bmatrix}
0.4625 & -0.0206 & -0.0197 & 0.4578 \\
-0.0198 & 0.0253 & 0.0235 & -0.0445 \\
0.0140 & 0.0083 & 0.0282 & -0.0485 \\
-0.0178 & 0.0109 & -0.0123 & 0.4839
\end{bmatrix} \hspace{1cm} (C3)$$

for the QST method 4, and

$$f(\rho'')_{\text{phys}} = \begin{bmatrix}
0.4766 & -0.0240 & -0.0347 & 0.4777 \\
-0.0012 & 0.0189 & 0.0150 & -0.0348 \\
0.0154 & 0.0072 & 0.0167 & -0.0455 \\
-0.0269 & -0.0014 & -0.0115 & 0.4879
\end{bmatrix} \hspace{1cm} (C4)$$

for the QST method 1, which are shown in Figs. 5(a) and 5(b), respectively.
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