Supplementary Information

Supplementary Figures

Supplementary Figure 1: Bloch sphere. Representation of the Bloch vector $|\psi_{ok}\rangle = \zeta|0\rangle + \xi|k\rangle$ on the Bloch sphere of the Hilbert sub-space spanned by the states $|0\rangle$ and $|k\rangle$ (where $k \in \{+, -\}$).
## Supplementary Tables

**Supplementary Table 1: Convention of basis operators.** List of full set of nine basis operators building a suitable basis for the decomposition of the 3D process matrix $\chi$ and the reduced 2D process matrix $\chi^{(+)}$ defined in Supplementary Equation (16).

| $E_m$ | Pauli operator | explicit expression | matrix representation | normalization factor $\alpha_m$ |
|-------|----------------|---------------------|-----------------------|-------------------------------|
| $E_1$ | $I_+ + I_-$   | $|+\rangle\langle+| + |\rangle\langle-|$ | $\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$ | $\sqrt{3}/2$ |
| $E_2$ | $\sigma^x_{+}$ | $|+\rangle\langle-| + |\rangle\langle+|$ | $\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$ | $\sqrt{3}/2$ |
| $E_3$ | $\sigma^y_{+}$ | $-i|+\rangle\langle-| + i|\rangle\langle+|$ | $\begin{pmatrix} 0 & 0 \\ 0 & -1 \end{pmatrix}$ | $\sqrt{3}/2$ |
| $E_4$ | $\sigma^z_{+}$ | $|+\rangle\langle+| - |\rangle\langle-|$ | $\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$ | $\sqrt{3}/2$ |
| $E_5$ | $\sigma^x_{+0}$ | $|+\rangle\langle0| + |0\rangle\langle+|$ | $\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$ | $\sqrt{3}/2$ |
| $E_6$ | $\sigma^y_{+0}$ | $-i|+\rangle\langle0| + i|0\rangle\langle+|$ | $\begin{pmatrix} 0 & 0 \\ 0 & -1 \end{pmatrix}$ | $\sqrt{3}/2$ |
| $E_7$ | $\sigma^x_{-0}$ | $|-\rangle\langle0| + |0\rangle\langle-|$ | $\begin{pmatrix} 0 & 1 \\ 0 & -1 \end{pmatrix}$ | $\sqrt{3}/2$ |
| $E_8$ | $\sigma^y_{-0}$ | $-i|-\rangle\langle0| + i|0\rangle\langle-|$ | $\begin{pmatrix} 0 & 0 \\ 0 & -1 \end{pmatrix}$ | $\sqrt{3}/2$ |
| $E_9$ | $I_0$         | $|0\rangle\langle0|$ | $\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$ | $\sqrt{3}$ |
**Supplementary Table 2: Pulse sequence for quantum process tomography.** List of the full set of nine basis states employed for QPT in this work. In the third column EXC means state initialization into the $m_s = 0$ state by optical pumping via a $\sim 4 \mu s$ long pulse of excitation light. The $(\tau)_{ijk}$ signify a microwave $i$-pulse of length $\tau$ on the $j$ to $k$ transition. DET means projective readout of the $m_s = 0$ population via $\sim 300$ ns of excitation light and simultaneous fluorescence detection.

| $\Psi_j$ | explicit expression | initialization | projective readout |
|----------|---------------------|----------------|-------------------|
| $\Psi_1$ | $\ket{+}$           | EXC + $\pi y_0^+$ | $\pi y_0^+ + \text{DET}$ |
| $\Psi_2$ | $\ket{-}$           | EXC + $\pi y_0^-$ | $\pi y_0^- + \text{DET}$ |
| $\Psi_3$ | $\ket{0}$           | EXC             | DET               |
| $\Psi_4$ | $\frac{1}{\sqrt{2}} (\ket{0} + \ket{+})$ | EXC + $(\frac{\pi}{2}) y_0^+$ | $(\frac{\pi}{2}) y_0^+ + \text{DET}$ |
| $\Psi_5$ | $\frac{1}{\sqrt{2}} (\ket{0} + i\ket{+})$ | EXC + $(\frac{\pi}{2}) x_0^+$ | $(\frac{\pi}{2}) x_0^+ + \text{DET}$ |
| $\Psi_6$ | $\frac{1}{\sqrt{2}} (\ket{0} + \ket{-})$ | EXC + $(\frac{\pi}{2}) y_0^-$ | $(\frac{\pi}{2}) y_0^- + \text{DET}$ |
| $\Psi_7$ | $\frac{1}{\sqrt{2}} (\ket{0} + i\ket{-})$ | EXC + $(\frac{\pi}{2}) x_0^-$ | $(\frac{\pi}{2}) x_0^- + \text{DET}$ |
| $\Psi_8$ | $\frac{1}{\sqrt{2}} (\ket{+} + \ket{-})$ | EXC + $(\frac{\pi}{2}) y_0^+ + \pi y_0^-$ | $\pi y_0^- + (\frac{\pi}{2}) y_0^+ + \text{DET}$ |
| $\Psi_9$ | $\frac{1}{\sqrt{2}} (\ket{+} + i\ket{-})$ | EXC + $(\frac{\pi}{2}) y_0^+ + \pi x_0^-$ | $\pi x_0^- + (\frac{\pi}{2}) y_0^+ + \text{DET}$ |

**Supplementary Table 3: Microwave pulse convention.** Definition of the rotation operators of the Bloch vector on the respective Hilbert sub-space spanned by either $\ket{0}$ and $\ket{+}$ or $\ket{0}$ and $\ket{-}$. The phase factor is acquired starting from the $\ket{0}$ state.

| MW pulse phase | pulse type | state transformation | pulse type | state transformation |
|---------------|------------|---------------------|------------|---------------------|
| $\sin(\omega_0 t)$ | $(\pi/2) x_0^+$ | $\begin{pmatrix} 1 & 0 & -1 \\ -1 & 0 & 1 \end{pmatrix} / \sqrt{2}$ | $(\pi/2) x_0^-$ | $\begin{pmatrix} \sqrt{2} & 0 & 0 \\ 0 & 1 & -1 \end{pmatrix} / \sqrt{2}$ |
| | $(\pi/2) x_0^+$ | $\begin{pmatrix} 0 & \sqrt{2} & 0 \\ -1 & 0 & 1 \end{pmatrix} / \sqrt{2}$ | $(\pi/2) x_0^+$ | $\begin{pmatrix} \sqrt{2} & 0 & 0 \\ 0 & 1 & 1 \end{pmatrix} / \sqrt{2}$ |
| | $(\pi/2) y_0^+$ | $\begin{pmatrix} 0 & \sqrt{2} & 0 \\ 1 & 0 & 1 \end{pmatrix} / \sqrt{2}$ | $(\pi/2) y_0^+$ | $\begin{pmatrix} \sqrt{2} & 0 & 0 \\ 0 & 1 & -1 \end{pmatrix} / \sqrt{2}$ |
| | $(\pi/2) y_0^+$ | $\begin{pmatrix} 0 & \sqrt{2} & 0 \\ -1 & 0 & 1 \end{pmatrix} / \sqrt{2}$ | $(\pi/2) y_0^+$ | $\begin{pmatrix} \sqrt{2} & 0 & 0 \\ 0 & 1 & 1 \end{pmatrix} / \sqrt{2}$ |
Supplementary Notes

Supplementary Note 1: Quantum process tomography

Quantum process tomography provides a means of determining the process matrix of an unknown quantum process acting on a quantum state \([3]\). It allows for the determination of the fidelity with which a specific quantum operation is performed experimentally in comparison to the theoretical, ideal process. Consider a general initial mixed quantum state

\[ \rho_{\text{in}} = \sum_k p_k |k\rangle \langle k| \]  

(where the states \(|k\rangle \in \mathcal{H}^d\), \(0 \leq p_k \leq 1\) and \(\sum_k p_k = 1\)). An arbitrary operation \(\mathcal{E}\) acting on that state

\[ \rho_{\text{in}} \rightarrow \mathcal{E}(\rho_{\text{in}}) = \rho_{\text{out}} \]  

can be described by a quantum process matrix \(\chi_{mn}\) generating the final state

\[ \rho_{\text{out}} = \mathcal{E}(\rho_{\text{in}}) = \sum_{mn} \chi_{mn} E_m \rho_{\text{in}} E_m^{\dagger} \]  

where \(\{E_m\} \in SU(d)\) represent a full set of orthogonal basis operators. The fidelity of the quantum process is then given by the overlap between the experimentally performed transformation \(\mathcal{E}\) and the theoretically ideal transformation \(\mathcal{U}\) as

\[ F(\mathcal{E}, \mathcal{U}) = \text{Tr}(\chi_{\text{exp}} \chi_{\text{theo}}) \]  

with the experimental and theoretical representation of the process matrix \(\chi_{\text{exp}}\) and \(\chi_{\text{theo}}\), respectively [1]. Since we reconstruct the fidelity of an intrinsically fault-tolerant quantum gate by means of standard QPT based on relatively vulnerable dynamical phase shifts, it is instructive to normalize the fidelity of the \(i\)-th quantum gate \(F_i\) obtained over standard QPT with respect to the fidelity of the QPT operation itself \(F_{\text{id}}\) (i.e. an “empty” QPT run without quantum gate). Thus we obtain the relative fidelities

\[ \tilde{F}_i(\mathcal{E}, \mathcal{U}) = \frac{F_i}{F_{\text{id}}} \]
as a sensible means for benchmarking the quantum gate performance achieved in this work.

Supplementary Note 2: Theoretical concept of the QPT procedure

Quantum process tomography is performed in $d^4$ runs, in each of which the system has to be initialized into a (quasi-)pure state $|\Psi_j\rangle \in \{|\Psi_1\rangle, |\Psi_2\rangle, ..., |\Psi_j\rangle\}$, where the $d^2$ states $|\Psi_j\rangle$ are chosen such that the corresponding density matrices $\rho_j = |\Psi_j\rangle\langle\Psi_j|$ form a basis for the space of matrices:

$$\rho = \sum_j q_j \rho_j = \sum_j q_j |\Psi_j\rangle\langle\Psi_j|.$$  \hfill (6)

Now, for each of the $d^4$ runs the unknown quantum process performs the transformation $\mathcal{E}$ on the full set of basis states $\rho_{\text{in}}^j$ ($j = 1, ..., 9$):

$$\rho_{\text{in}}^j \rightarrow \mathcal{E}(\rho_{\text{in}}^j) = \rho_{\text{out}}^j.$$ \hfill (7)

In order to find an expression for the unknown transformation characterized by the process matrix $\chi_{mn}$ we decompose both $\mathcal{E}(\rho_{\text{in}}^j)$ and $E_m \rho_{\text{in}}^j E_n^\dagger$ in the chosen state basis $\rho_k = |\Psi_k\rangle\langle\Psi_k|:

$$\mathcal{E}(\rho_{\text{in}}^j) = \sum_k \lambda_{jk} \rho_k,$$ \hfill (8)

$$E_m \rho_{\text{in}}^j E_n^\dagger = \sum_k \beta_{jk}^{mn} \rho_k.$$ \hfill (9)

In general, the coefficients $\lambda_{jk}$ and $\beta_{jk}^{mn}$ are complex. While the $\beta_{jk}^{mn}$ are to be determined theoretically on the basis of the formerly defined set of $E_m$ and $\Psi_j$, the $\lambda_{jk}$ are reconstructed from experimental results. We can construct the theoretical and experimental $\lambda_{jk}$ by solving the linear system of equations for the observable

$$O_{pj} = \langle \Psi_p | \mathcal{E}(\rho_{\text{in}}^j) | \Psi_p \rangle = \sum_k \lambda_{jk} \langle \Psi_p | \rho_k | \Psi_p \rangle = \sum_k \lambda_{jk} |\langle \Psi_p | \Psi_k \rangle|^2.$$ \hfill (10)
corresponding to the $d^2$ projective measurements on the complete set of projection bases states $\Psi_p$. Analogously, we can determine the $\beta_{jk}^{mn}$ by solving the respective linear system of equations

$$\langle \Psi_p|E_mE^\dagger_n|\Psi_p\rangle = \sum_k \beta_{jk}^{mn} \langle \Psi_p|\rho_k|\Psi_p\rangle = \sum_k \beta_{jk}^{mn} |\langle \Psi_p|\Psi_k\rangle|^2.$$  \hspace{2cm} (11)

Combining both Supplementary Equation (8) and Supplementary Equation (9) we obtain

$$\mathcal{E}(\rho_j^{\text{in}}) = \sum_{mn} \chi_{mn} E_mE^\dagger_n = \sum_{mn} \chi_{mn} \sum_k \beta_{jk}^{mn} \rho_k = \sum_k \lambda_{jk} \rho_k.$$ \hspace{2cm} (12)

In fact, this relation holds for each $\rho_k$ separately, so we may write

$$\sum_{mn} \chi_{mn} \beta_{jk}^{mn} = \lambda_{jk}. \hspace{2cm} (13)$$

If we now compute for every $\rho_j$ the components $\kappa_{jk}^{mn}$ as the generalized inverse of $\beta_{jk}^{mn}$

$$\sum_{mn} \chi_{mn} \sum_{jk} \beta_{jk}^{mn} \kappa_{pq}^{jk} = \sum_j \kappa_{jk}^{pq} \lambda_{jk} = \delta_{mp} \delta_{nq}.$$ \hspace{2cm} (14)

we can find an explicit expression for the process matrix

$$\chi_{mn} = \sum_{jk} \kappa_{jk}^{mn} \lambda_{jk} \hspace{2cm} (15)$$

given in the basis of the generators $E_m, E_n$.

**Supplementary Note 3: Theoretical and experimental choice of basis states and projection operators**

For the quantum process tomography the basis operators $E_k$ have to be chosen suitably. In a minimal setting these ought to be a full set of 8 generators of the $\mathcal{SU}(3)$, the so called the Gell-Mann matrices. These matrices need to be Hermitian, traceless ($\text{Tr}(E_i) = 0$) and satisfy $\text{Tr}(E_i E_j) = 2\delta_{ij}$. For the NV$^- \text{ ground state triplet}$ the suggested set is presented in Supplementary Table 1. Here, we suggest a set of nine generators $E_m$ of the $\mathcal{U}(3)$, as in this representation
the $4 \times 4$ process matrix $\chi^{(+)}$ acting only in the Hilbert subspace $\mathcal{H}^2$ spanned by the computational states $|+\rangle$ and $|-\rangle$ can be immediately extracted from the full $9 \times 9$ process matrix $\chi$ as

$$\chi^{(+)} = \left( \text{Tr}\{\hat{\chi}^{(+)}\} \right)^{-1} \cdot \hat{\chi}^{(+)} .$$

(16)

with

$$\hat{\chi}^{(+)} = \sum_{m,n=1}^{4} \text{Tr}\{|e_m\langle e_n|\chi_{mn}\} \cdot |e_m\rangle\langle e_n|$$

(17)

where the $|e_m\rangle = (0, 0, ..., \delta_{im}, ..., 0) \in \mathbb{R}^4$ are unit vectors. The operators $E_2$ to $E_8$ are effectively Pauli operators acting only on a two-dimensional subspace of the total state space. The first and the ninth generator are chosen as (a suitable linear combination of) sub-space identity operators $I_+, I_-$ and $I_0$.

The respective set of proposed basis states $\Psi_j$ is given in Supplementary Table 2. These states of the NV− ground state triplet are to be prepared on the basis of microwave Rabi pulses applied to the $|0\rangle$ to $|+\rangle$ and the $|0\rangle$ to $|-\rangle$ transitions inducing rotations of the Bloch vectors on the two coupled Bloch sub-spheres spanned by the states $|0\rangle \& |+\rangle$ and $|0\rangle \& |-\rangle$, respectively (cf. Supplementary Figure 1). An initial Bloch vector $|\psi\rangle$ is rotated to state $|\psi'\rangle$ upon application of a Rabi pulse $R_{0k}$ following $|\psi'\rangle_{0k} = R_{0k}|\psi\rangle_{0k}$, where $|\psi\rangle_{0k} \in \mathcal{H}^2 = \{\zeta|0\rangle + \xi|k\rangle \mid \zeta, \xi \in \mathbb{C}\}$. Here, we want to employ the following convention for dynamic phase shifts induced by resonant microwave pulses of length $\tau = \beta / \Omega(t)$ on either of the $|0\rangle \leftrightarrow |+\rangle$ and the $|0\rangle \leftrightarrow |-\rangle$ transition:

$$R_{0+}(\alpha, \beta) = \begin{pmatrix}
\cos(\beta/2) & 0 & -ie^{-i\alpha}\sin(\beta/2) \\
0 & 1 & 0 \\
-ie^{i\alpha}\sin(\beta/2) & 0 & \cos(\beta/2)
\end{pmatrix}$$

(18)

$$R_{0-}(\alpha, \beta) = \begin{pmatrix}
1 & 0 & 0 \\
0 & \cos(\beta/2) & -e^{-i\alpha}\sin(\beta/2) \\
0 & e^{i\alpha}\sin(\beta/2) & \cos(\beta/2)
\end{pmatrix}$$

(19)

where $\beta$ determines the rotation angle and $\alpha$ the rotation axis. An $x$ pulse is considered a microwave sine pulse, an $\bar{x}$ pulse has a relative phase shift of $\alpha = +\pi$, a $y$ pulse a phase shift of $\alpha = +\frac{\pi}{2}$, and a $\bar{y}$ pulse a phase shift of $\alpha = -\frac{\pi}{2}$. Explicit expressions for the rotation matrices of
the respective pulses are shown in Supplementary Table 3. These sub-space rotation operations can be conveniently visualized on two coupled sub-space Bloch spheres. Supplementary Figure 1 shows the sub-space Bloch sphere spanned by the states $|0\rangle$ and $|+\rangle$. Bear in mind that as the two sub-space Bloch spheres are coupled, global phase factors arising due to rotations on a sub-space Bloch sphere are not negligible; indeed, they are relative phase factors on the respective generalized eight-dimensional Bloch sphere comprising both sub-space Bloch spheres.

The corresponding experimental preparation routines for the set of basis states $\psi_j$ are proposed along with their corresponding projective readout sequences in the third and forth column of Supplementary Table 2, respectively. In the case of the NV$^-$ centre the state initialization is always performed by optical pumping into the $|0\rangle$ state followed by a suitable combination of microwave $\pi$ and $\pi/2$ pulses. The projective readout follows the reverse scheme projecting the obtained state $\rho^\text{out}$ back onto the $|0\rangle$ basis. This allows for the reconstructed of the decomposition of $\rho^\text{out}$ in the basis of $\rho_j$.

Supplementary Note 4: Maximum likelihood estimation procedure

In order to extract the experimentally obtained process fidelity we fit the experimental data with a proper theoretical model by means of a maximum likelihood estimation (MLE) procedure. For this purpose the maximum likelihood function

$$f(\vec{q}) = \sum_{j,p} \left( \langle \psi_p | E_j \rho_j | \psi_p \rangle - \sum_{m,n} \chi_{mn}(\vec{q}) \langle \psi_p | E_m | \psi_j \rangle \langle \psi_j | E_n | \psi_p \rangle \right)^2 - \Lambda \left( \sum_{m,n,r} \chi_{mn}(\vec{q}) \text{Tr}(\alpha_m \alpha_r \alpha_n E_mE_n) - \delta_{r,1} \right)$$

is to be minimized, where $\Lambda$ is a Lagrange multiplier and $\delta_{r,1}$ the Kronecker delta. The $\alpha_m$ are normalization factors (see Supplementary Table 1) that allow for an direct extraction of the process matrix $\chi^{(+)} \in \mathcal{H}^2$ (defined on the computational state space) from the fitted process.
The matrix $\chi^{(+0)} \in \mathcal{H}^3$ (defined on the total system space) as stated in Supplementary Equation (16). The first term in Supplementary Equation (20) ensures hermiticity, while the second term sets the degree of positivity by means of a Lagrange multiplier $\Lambda$. Trace-preservation of the process matrix is satisfied per constructionem by the parametrized representation

$$\chi(q) = \frac{Q^\dagger(q)Q(q)}{\text{Tr}\{Q^\dagger(q)Q(q)\}}$$

or element-wise

$$\chi_{mn}(q) = \left( \sum_{m',n'} \delta_{m',n'} (Q_{km'})^* Q_{kn'} \right)^{-1} \cdot \left( \sum_k (Q_{km})^* Q_{kn} \right)$$

where the complex valued matrix $Q(q)$ is a triangular matrix parametrized by a real valued vector $q$ containing $d^4 - d^2$ elements. For the present case of $d = 3$ the $Q(q)$ matrix can be written as

$$Q(q) = \begin{pmatrix}
q_1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
q_{2+10} & q_{18} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
q_3+iq_{11} & q_{19}+iq_{26} & q_{23} & 0 & 0 & 0 & 0 & 0 & 0 \\
q_4+iq_{12} & q_{20}+iq_{27} & q_{34}+iq_{40} & q_{46} & 0 & 0 & 0 & 0 & 0 \\
q_{5+i13} & q_{21}+iq_{28} & q_{35}+iq_{41} & q_{47}+iq_{52} & q_{57} & 0 & 0 & 0 & 0 \\
q_6+iq_{14} & q_{22}+iq_{29} & q_{36}+iq_{42} & q_{48}+iq_{53} & q_{58}+iq_{62} & q_{66} & 0 & 0 & 0 \\
q_7+iq_{15} & q_{23}+iq_{30} & q_{37}+iq_{44} & q_{49}+iq_{54} & q_{59}+iq_{63} & q_{67}+iq_{70} & q_{73} & 0 & 0 \\
q_{8+i16} & q_{24}+iq_{31} & q_{38}+iq_{45} & q_{51}+iq_{56} & q_{51}+iq_{65} & q_{69}+iq_{72} & q_{75}+iq_{77} & q_{79}+iq_{80} & q_{81}
\end{pmatrix}$$

where $q = (q_1, q_2, q_3, \ldots, q_{81})$ denotes the set of 81 fit parameters. In principle a suitable set of start values $\tilde{q}_0$ for the iterative fit could be extracted from $\tilde{\chi}_{\text{exp}}$ obtained from the raw experimental data. Practically, however, in the case of $d > 2$ the reverse element-wise dependence of Supplementary Equation (22), i.e. $Q_{kn}$ as a function of the $\tilde{\chi}_{mn}^{\text{exp}}$ is non-trivial:

$$Q_{kn}(\tilde{\chi}_{mn}^{\text{exp}}) = \tilde{q}_0.$$
Supplementary Equation (24) is highly non-trivial to compute. Thus, in this work we chose to find a start parameter set \( \vec{q}_0 \) as follows: First, we approximate the experimentally obtained \( \tilde{\chi}_{mn}^{\text{exp}} \) by a positive definite version of that matrix through substitution of the (anyway small) negative diagonal entries in the diagonalized form of \( \vec{q}_0(\tilde{\chi}_{mn}^{\text{exp}}) \) by suitably small, but positive values) and subsequently performing a Cholesky decomposition. From the lower unit triangular matrix of the Cholesky decomposition we can extract a start value set \( \vec{q}_0 \) for the maximum likelihood estimation procedure.

**Supplementary Note 5: Data Evaluation and error estimation for maximum likelihood fit of \( \chi \) and \( F \)**

Due to the complex relation between the measured data and the final process matrix \( \chi \) and the fidelity \( F \) the error calculation is non-trivial. Therefore, a Monte Carlo based error estimation method is employed here. The observables, i.e. the projective readout data from Supplementary Equation (10) are fitted by Gaussian distributions and the mean value \( \bar{O}_{pj} \) and standard deviation \( \sigma_{O_{pj}} \) of each observable fit is extracted. From a normally distributed set of Monte Carlo sampled observable values we obtain the corresponding distribution for the \( \lambda_{jk} \) coefficients and can extract mean value \( \bar{\lambda}_{jk} \) and standard deviation \( \sigma_{\lambda_{jk}} \) for each of the 81 \( \lambda_{jk} \). From a normally distributed Monte Carlo sampled set of each of the 81 \( \lambda_{jk} \) coefficients we obtain a distribution for the \( \chi_{mn}^{\text{raw}} \) elements and can extract 81 mean values \( \bar{\chi}_{mn}^{\text{raw}} \) and standard deviations \( \sigma_{\chi_{mn}^{\text{raw}}} \) as well as a mean process matrix

\[
\chi^{\text{raw}} = \sum_{m,n=1}^{9} \text{Tr}\{|e_m\rangle\langle e_n|\chi^{\text{raw}}_{mn}\} \cdot |e_m\rangle\langle e_n|
\]

(25)

where the \( |e_m\rangle = (0, 0, ..., \delta_{im}, ..., 0) \in \mathbb{R}^9 \) are unit vectors.

Now the raw data process matrix \( \chi^{\text{raw}} \) is subject to the MLE procedure described above (where the start parameter set is derived from the mean process matrix: \( \chi(\vec{q}_0(\chi^{\text{raw}})) \)) delivering
a fitted process matrix $\chi^{\text{MLE}}$. Ultimately, the total process fidelity of the $i$-th gate is computed with respect to the MLE fitted process matrix $\chi_i^{\text{MLE}}$ as

$$F_i = \text{Tr}(\chi_i^{\text{MLE}} \chi_i^{\text{theo}})$$

providing a reasonable figure of merit for the achieved performance of the quantum gate of interest.

In order to obtain an error estimation for this MLE fitted process matrix and the fidelity a large sample of normally distributed $\chi_{\text{raw MC}}$ with the previously determined standard deviation $\sigma_{\chi_{\text{raw}}}$ is subject to the maximum likelihood procedure as well resulting in a non-Gaussian distribution for the $\chi_{\text{MLE MC}}$. The error of the fidelity was derived from the (in general asymmetric) distribution of the fidelities computed from the respective Monte Carlo sets $\chi_{\text{MLE MC}}$. The error bars given in the main article cover a 68.3% confidence interval around the fidelity values $F_i$ and have to be seen as an upper bound error estimate, as the Monte Carlo sampled sets of $\chi_{\text{raw MC}}$ elements were uncorrelated. The obtained fidelity errors are in good agreement with the residua between the fidelities computed from the unfitted $\chi_{\text{raw}}$ and the fidelity computed from the fitted $\chi^{\text{MLE}}$.

In order to remove the fidelity bias originating from the infidelity of the characterizing quantum process tomography itself, the extracted fidelities of the $i$-th holonomic quantum gate $\tilde{F}_i$ were obtained from a normalization of the respective total process fidelity $F_i$ by the fidelity of the identity gate $F_{\text{ID}}$: $\tilde{F}_i = \frac{F_i}{F_{\text{ID}}}$.

**Supplementary References**

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[2] James, D. F. V., Kwiat, P. G., Munro, W. J. & White, A. G. Measurement of qubits. *Phys. Rev. A.*, 64, 5, 052312 (2001).

[3] Nielsen, M. A. & Chuang, I. L. *Quantum Computation and Quantum Information*. (Cambridge University Press, Cambridge, 2005).