PROOF OF MATHEMATICAL STATEMENTS

Lemma 1. *If X is an instrumental variable for a potential outcome model (A, B, PO), then*

$$\Pr\left(B_{\mathrm{do}(A=a)} = b\right) \geq \sup_{x\in\mathcal{X}} \Pr\left(A = a, B = b \mid X = x\right). \tag{1}$$

Moreover, if A is binary (i.e., $\mathcal{A} = \{0, 1\}$), then

$$\Pr\left(B_{\mathrm{do}(A=0)} = b_0,\ B_{\mathrm{do}(A=1)} = b_1\right) \leq \inf_{x\in\mathcal{X}} \Big[\Pr\left(A = 0, B = b_0 \mid X = x\right) + \Pr\left(A = 1, B = b_1 \mid X = x\right)\Big]. \tag{2}$$

Proof. For the first inequality, note that for any $x \in \mathcal{X}$ we have

$$\begin{aligned}\Pr\left(B_{\mathrm{do}(A=a)} = b\right) &\overset{\text{(i)}}{=} \Pr\left(B_{\mathrm{do}(A=a)} = b \mid X = x\right)\\ &\overset{\text{(ii)}}{\geq} \Pr\left(A = a, B_{\mathrm{do}(A=a)} = b \mid X = x\right)\\ &\overset{\text{(iii)}}{=} \Pr\left(A = a, B = b \mid X = x\right),\end{aligned}$$

where (i) follows from the independence of X and the potential outcomes, (ii) uses the fact that the marginal probability dominates the corresponding joint probability, and (iii) follows from the do-calculus rule (see, e.g., Ref. [1]),

$$\Pr(B_{\mathrm{do}(A=a)} = b \mid X, A = a) = \Pr(B = b \mid X, A = a), \tag{3}$$

together with Bayes rule the assumption that X does not alter the potential outcomes, i.e., $B_{\mathrm{do}(A=a,X=x)} = B_{\mathrm{do}(A=a,X=x')} \equiv B_{\mathrm{do}(A=a)}$. Taking the supremum over $x \in \mathcal{X}$ proves the first part.

For the second inequality, assume that A is binary. Then, for any fixed $x \in \mathcal{X}$, we can write

$$\begin{aligned}\Pr\left(B_{\mathrm{do}(A=0)} = b_0,\ B_{\mathrm{do}(A=1)} = b_1\right) &\overset{\text{(v)}}{=} \Pr\left(B_{\mathrm{do}(A=0)} = b_0,\ B_{\mathrm{do}(A=1)} = b_1 \mid X = x\right)\\ &= \Pr\left(A = 0, B_{\mathrm{do}(A=0)} = b_0,\ B_{\mathrm{do}(A=1)} = b_1 \mid X = x\right)\\ &\quad + \Pr\left(A = 1, B_{\mathrm{do}(A=0)} = b_0,\ B_{\mathrm{do}(A=1)} = b_1 \mid X = x\right)\\ &\overset{\text{(vi)}}{\leq} \Pr\left(A = 0, B_{\mathrm{do}(A=0)} = b_0 \mid X = x\right)\\ &\quad + \Pr\left(A = 1, B_{\mathrm{do}(A=1)} = b_1 \mid X = x\right)\\ &= \Pr\left(A = 0, B = b_0 \mid X = x\right) + \Pr\left(A = 1, B = b_1 \mid X = x\right),\end{aligned}$$

where (v) is justified by the joint independence $X \perp\!\!\!\perp (B_{\mathrm{do}(A=0)}, B_{\mathrm{do}(A=1)})$, and the inequality (vi) follows from the properties of probabilities. Taking the infimum over $x \in \mathcal{X}$ completes the proof. ∎

Lemma 2. *Let X be an instrumental variable for a potential outcome model (A, B, PO), with A and B binary. Then*

$$\sum_{b_0, b_1\in\mathcal{B}} \inf_{x\in\mathcal{X}} \sum_{a\in\{0,1\}} \Pr(A = a, B = b_a \mid X = x) = 2 - \frac{\mathrm{CHSH}' + \mathrm{CHSH}''}{4}, \tag{4}$$

where the right-hand side involves the sum of two CHSH scores,

$$\text{CHSH} = \langle \hat{A}_x \hat{B}_y \rangle + \langle \hat{A}_{x'} \hat{B}_y \rangle + \langle \hat{A}_x \hat{B}_{y'} \rangle - \langle \hat{A}_{x'} \hat{B}_{y'} \rangle, \tag{5}$$

and equivalent ones using symmetry transformations.

Proof. Define the variables $\hat{A}_X$ (and $\hat{B}_Y$) by the rule that they assume value +1 if $A_{\text{do}(X)} = 0$ ($B_{\text{do}(Y)} = 0$, respectively) and −1 otherwise. Then, write Bell's probabilities as in

$$\Pr(AB \mid XY)_{\text{Bell}} = \frac{1}{4}\left[1 + (-1)^A \langle \hat{A}_X \rangle + (-1)^B \langle \hat{B}_Y \rangle + (-1)^{A+B} \langle \hat{A}_X \hat{B}_Y \rangle\right]. \tag{6}$$

Define

$$\begin{aligned}
\mathcal{I} &= \sum_{b_0, b_1 \in \mathcal{B}} \inf_{x \in \mathcal{X}} \sum_{a \in \{0,1\}} \Pr(A = a, B = b_a \mid X = x) \\
&= \sum_{b_0, b_1 \in \mathcal{B}} \Big[\Pr(A = 0, B = b_0 \mid X = f(b_0, b_1)) + \Pr(A = 1, B = b_1 \mid X = f(b_0, b_1)) \Big],
\end{aligned} \tag{7}$$

for some function $f : \mathcal{B} \times \mathcal{B} \to \mathcal{X}$. Let $x_{b_0 b_1} = f(b_0, b_1)$. Since B is binary, we can expand

$$\begin{aligned}
\mathcal{I} = \Pr(A = 0, B = 0 \mid X = x_{00}) &+ \Pr(A = 1, B = 0 \mid X = x_{00}) \\
+ \Pr(A = 0, B = 1 \mid X = x_{11}) &+ \Pr(A = 1, B = 1 \mid X = x_{11}) \\
+ \Pr(A = 0, B = 0 \mid X = x_{01}) &+ \Pr(A = 1, B = 1 \mid X = x_{01}) \\
+ \Pr(A = 0, B = 1 \mid X = x_{10}) &+ \Pr(A = 1, B = 0 \mid X = x_{10}).
\end{aligned} \tag{8}$$

Now remember that $\Pr(AB \mid X) = \Pr(AB \mid X, Y = A)_{\text{Bell}}$ and use it to substitute Eq. (6) into the above expression. By doing so, all single-party terms cancel, giving

$$\begin{aligned}
\mathcal{I} = 1 &+ \frac{-\langle \hat{A}_{x_{00}} \hat{B}_0 \rangle + \langle \hat{A}_{x_{00}} \hat{B}_1 \rangle - \langle \hat{A}_{x_{11}} \hat{B}_0 \rangle - \langle \hat{A}_{x_{11}} \hat{B}_1 \rangle}{4} \\
&+ 1 + \frac{\langle \hat{A}_{x_{01}} \hat{B}_0 \rangle - \langle \hat{A}_{x_{01}} \hat{B}_1 \rangle + \langle \hat{A}_{x_{10}} \hat{B}_0 \rangle + \langle \hat{A}_{x_{10}} \hat{B}_1 \rangle}{4},
\end{aligned} \tag{9}$$

which is precisely Eq. (4). ■

Proposition 3. *If a TPM process has no quantum memory, then*

$$W = \sum_{\lambda} \mu_\lambda \, \eta^{\lambda}_{\mathbf{A}'} \otimes N^{\lambda}_{\mathbf{A} \to \mathbf{B}'}, \tag{10}$$

*where $\mu_\lambda \geq 0$ with $\sum_\lambda \mu_\lambda = 1$, each $\eta^{\lambda}_{\mathbf{A}'}$ is a density operator, and $N^{\lambda}_{\mathbf{A} \to \mathbf{B}}$ is the Choi–Jamiołkowski representation of a channel from **A** to **B**. Moreover, quantum TPM correlations of the form of*

$$\Pr(AB \mid X) = \operatorname{Tr}[(E_{A|X} \otimes \rho_A \otimes F_B) W] \tag{11}$$

and

$$\Pr(B_{\text{do}(A=a)} = b) = \operatorname{Tr}[(\text{id}_{\mathbf{A}'} \otimes \rho_{A=a} \otimes F_{B=b}) W] \tag{12}$$

are compatible with a classical causal model.

Proof. If a TPM process has no quantum memory, then the initial state $\varrho_{\mathbf{A'E}}$ is mapped as $\varrho_{\mathbf{A'E}} \mapsto \mathcal{M}^{\mathrm{EB}}_{\mathbf{E}\to\mathbf{E}}(\varrho_{\mathbf{A'E}})$, where $\mathcal{M}^{\mathrm{EB}}_{\mathbf{E}\to\mathbf{E}}$ is an entanglement-breaking channel. Hence,

$$\mathcal{M}^{\mathrm{EB}}_{\mathbf{E}\to\mathbf{E}}(\varrho_{\mathbf{A'E}}) = \sum_\lambda \mu_\lambda\, \varrho^\lambda_{\mathbf{A'}} \otimes \varrho^\lambda_{\mathbf{E}}. \tag{13}$$

The TPM process operator is then

$$\begin{aligned} W_{\mathrm{CM}} &= \mathrm{Tr}_{\mathbf{EE'}}\Big[\big(\mathcal{M}^{\mathrm{EB}}_{\mathbf{E}\to\mathbf{E}}(\varrho_{\mathbf{A'E}}) \otimes \mathrm{id}_{\mathbf{ABE'}}\big)|U\rangle\!\rangle\langle\!\langle U|\Big] \\ &\overset{(*)}{=} \mathrm{Tr}_{\mathbf{E}}\Big[\big(\mathcal{M}^{\mathrm{EB}}_{\mathbf{E}\to\mathbf{E}}(\varrho_{\mathbf{A'E}}) \otimes \mathrm{id}_{\mathbf{AB}}\big)K_{\mathbf{AE}\to\mathbf{B}}\Big] \\ &= \sum_\lambda \mu_\lambda\, \varrho^\lambda_{\mathbf{A'}} \otimes \mathrm{Tr}_{\mathbf{E}}\big[(\varrho^\lambda_{\mathbf{E}} \otimes \mathrm{id}_{\mathbf{AB}})K_{\mathbf{AE}\to\mathbf{B}}\big], \end{aligned} \tag{14}$$

where in $(*)$ we have defined $K_{\mathbf{AE}\to\mathbf{B}} = \mathrm{Tr}_{\mathbf{E'}}|U\rangle\!\rangle\langle\!\langle U|$. Next, define

$$N^\lambda_{\mathbf{A}\to\mathbf{B}} = \mathrm{Tr}_{\mathbf{E}}\big[(\varrho^\lambda_{\mathbf{E}} \otimes \mathrm{id}_{\mathbf{AB}})K_{\mathbf{AE}\to\mathbf{B}}\big]. \tag{15}$$

For each λ, we have $N^\lambda_{\mathbf{A}\to\mathbf{B}} \succcurlyeq 0$ and

$$\begin{aligned} \mathrm{Tr}_{\mathbf{B}}\, N^\lambda_{\mathbf{A}\to\mathbf{B}} &= \mathrm{Tr}_{\mathbf{E}}\Big[(\varrho^\lambda_{\mathbf{E}} \otimes \mathrm{id}_{\mathbf{AB}})\mathrm{Tr}_{\mathbf{B}}\, K_{\mathbf{AE}\to\mathbf{B}}\Big] \\ &= \mathrm{id}_{\mathbf{A}}\, \mathrm{Tr}\,\varrho^\lambda_{\mathbf{E}} \\ &= \mathrm{id}_{\mathbf{A}}, \end{aligned} \tag{16}$$

since $\mathrm{Tr}_{\mathbf{B}}\, K_{\mathbf{AE}\to\mathbf{B}} = \mathrm{id}_{\mathbf{AE}}$. Thus, each $N^\lambda_{\mathbf{A}\to\mathbf{B}}$ is indeed the CJ operator of a channel $\mathbf{A} \to \mathbf{B}$.

Finally, quantum TPM probabilities take the form

$$\begin{aligned} \Pr(AB \mid X) &= \mathrm{Tr}\big[(E_{A|X} \otimes \rho_A \otimes F_B)\, W\big] \\ &= \sum_\lambda \mu_\lambda\, \mathrm{Tr}\big(E_{A|X}\, \eta^\lambda_{\mathbf{A'}}\big)\, \mathrm{Tr}\big[(\rho_A \otimes F_B)\, N^\lambda_{\mathbf{A}\to\mathbf{B}}\big]. \end{aligned} \tag{17}$$

This naturally suggests the definitions $\Pr(\Lambda = \lambda) = \mu_\lambda$, $\Pr(A \mid X, \Lambda = \lambda) = \mathrm{Tr}(E_{A|X}\, \eta^\lambda_{\mathbf{A'}})$, $\Pr(B \mid A, \Lambda = \lambda) = \mathrm{Tr}[(\rho_A \otimes F_B)\, N^\lambda_{\mathbf{A}\to\mathbf{B}}]$. These are valid probability distributions. They are obviously positive and the normalisation of $\Pr(B \mid A, \Lambda = \lambda)$ follows from the fact that each $N^\lambda_{\mathbf{A}\to\mathbf{B}}$ is the CJ operator of a channel:

$$\begin{aligned} \sum_b \Pr(B = b \mid A, \Lambda = \lambda) &= \mathrm{Tr}\big[(\rho_A \otimes \mathrm{id}_{\mathbf{B}})\, N^\lambda_{\mathbf{A}\to\mathbf{B}}\big] \\ &= \mathrm{Tr}\Big(\rho_A\, \mathrm{Tr}_{\mathbf{B}} N^\lambda_{\mathbf{A}\to\mathbf{B}}\Big) \\ &= 1, \end{aligned} \tag{18}$$

which works only because $\mathrm{Tr}_{\mathbf{B}}\, N^\lambda_{\mathbf{A}\to\mathbf{B}} = \mathrm{id}_{\mathbf{A}}$. Quantum do-probabilities in turn read

$$\begin{aligned} \Pr(B_{\mathrm{do}(A=a)} = b) &= \mathrm{Tr}\,[(\mathrm{id}_{\mathbf{A'}} \otimes \rho_{A=a} \otimes F_{B=b})W] \\ &= \sum_\lambda \mu_\lambda \mathrm{Tr}\,[(\rho_{A=a} \otimes F_{B=b})N^\lambda_{\mathbf{A}\to\mathbf{B}}] \\ &= \sum_\lambda \Pr(\Lambda = \lambda)\, \Pr(B = b \mid A = a, \Lambda = \lambda). \end{aligned} \tag{19}$$

This concluses the proof. ■

Proposition 4. *Any TPM process, characterised by a process operator $W(\varrho, U)$ and satisfying any of the conditions (C1), (C2), or (C3) discussed above, is compatible with classical causal models and thus does not violate any classical inequality.*

Proof. The fact that condition (C1) implies classicality was already established in Proposition 3. Next, invoking the quantum analogue of the mapping in Eq. (**??**),

$$\Pr(AB \mid X) = \mathrm{Tr}\,[(E_{A|X} \otimes G_{B|A})\,\varrho], \tag{20}$$

it follows directly that the joint measurability of either $E_{A|X}$ or $G_{B|A}$ entails classicality. This likewise yields the same conclusion for CC-type process operators, for which $G_{B|A}$ is independent of A. Finally, the convex hull of DC-type process operators coincides with the set of processes possessing no quantum memory, which, by Proposition 3, are classical. By convexity, it follows that (C3) also implies classicality. ■

Proposition 5. *Consider the measurement assemblages $G^{\alpha}_{B|A}$ generated through the α-partial* SWAP *gate,*

$$U_{\mathrm{PS}}(\alpha) = \cos(\alpha/2)\mathrm{id} + \mathrm{i}\sin(\alpha/2)U_{\textit{SWAP}}, \tag{21}$$

via

$$G^{\alpha}_{B|A} = \mathrm{Tr}_{\mathrm{A}}[U_{\mathrm{PS}}(\alpha)^{\dagger}(\mathrm{id}_{\mathbf{E}'} \otimes F_B)U_{\mathrm{PS}}(\alpha)(\mathrm{id}_{\mathbf{E}} \otimes \rho_A)] \tag{22}$$

If A and B are binary variables, $G^{\alpha}_{B|A}$ is always jointly measurable for $0 \leq \alpha \leq \pi/2$ and $\alpha = \pi$ and can be incompatible otherwise.

Proof. First, note that with some manipulations one can write

$$G^{\alpha}_{B|A} = \cos^2\left(\frac{\alpha}{2}\right)\mathrm{Tr}\,(F_B\sigma_A)\mathrm{id} + \sin^2\left(\frac{\alpha}{2}\right)F_B - \mathrm{i}\sin\left(\frac{\alpha}{2}\right)\cos\left(\frac{\alpha}{2}\right)[F_B, \sigma_A]. \tag{23}$$

Joint measurability for $\alpha = \pi$ is trivial, since $F^{\pi}_{B|A} = F_B$, i.e., it does not depend on A. Eq. (23) is linear in both states and POVM effects and thus we can restrict ourselves to projective measurements and pure states. Moreover, we explore the fact that $U_{\mathrm{PS}}(\alpha)$ lies within the symmetric subspace to set, without loss of generality, $\sigma_{A=0} = |0\rangle\langle 0|$ and $\sigma_{A=1} = |\theta_{\mathrm{S}}, \phi_{\mathrm{S}}\rangle\langle\theta_{\mathrm{S}}, \phi_{\mathrm{S}}|$ as well as $E_{B=0} = |\theta_{\mathrm{E}}, \phi_{\mathrm{E}}\rangle\langle\theta_{\mathrm{E}}, \phi_{\mathrm{E}}|$. Moreover, we parametrise the POVM effects as follows

$$F^{\alpha}_{B=0|A} = \frac{1}{2}[(1+\gamma_A)\mathrm{id} + \boldsymbol{r}_A \cdot \boldsymbol{\sigma}], \tag{24}$$

where $\boldsymbol{r}_A \cdot \boldsymbol{\sigma} = r^x_A\sigma_x + r^y_A\sigma_y + r^z_A\sigma_z$. We do so in order to apply the criterion for compatibility of a pair of qubit POVMs (see Ref. [2] and references therein). The criterion states that $F^{\alpha}_{B|A}$ is jointly measurable if and only if

$$(\boldsymbol{r}_0 \cdot \boldsymbol{r}_1 - \gamma_0\gamma_1)^2 - \left(1 - \mathcal{F}_0^2 - \mathcal{F}_1^2\right)\left(1 - \frac{\gamma_0^2}{\mathcal{F}_0^2} - \frac{\gamma_1^2}{\mathcal{F}_1^2}\right) \geq 0, \tag{25}$$

where

$$\mathcal{F}_a = \frac{1}{2}\left(\sqrt{(1+\gamma_a)^2 - \|\boldsymbol{r}_a\|^2} + \sqrt{(1-\gamma_a)^2 - \|\boldsymbol{r}_a\|^2}\right). \tag{26}$$

A routine calculation shows that for $A = 0$ one has

$$\begin{aligned} y_0 &= \cos^2\left(\frac{\alpha}{2}\right)\cos\theta_{\mathrm{E}}, \\ x_0 &= \sin\left(\frac{\alpha}{2}\right)\sin\theta_{\mathrm{E}}\sin\left(\frac{\alpha}{2}+\phi_{\mathrm{E}}\right), \\ y_0 &= -\sin\left(\frac{\alpha}{2}\right)\sin\theta_{\mathrm{E}}\cos\left(\frac{\alpha}{2}+\phi_{\mathrm{E}}\right), \\ z_0 &= \sin^2\left(\frac{\alpha}{2}\right)\cos\theta_{\mathrm{E}}, \end{aligned}$$

and, similarly, for $A = 1$,

$$\begin{aligned} y_1 &= \cos^2\left(\frac{\alpha}{2}\right)\left[\cos\theta_{\mathrm{E}}\cos\theta_{\mathrm{S}} + \cos(\phi_{\mathrm{S}}-\phi_{\mathrm{E}})\sin\theta_{\mathrm{E}}\sin\theta_{\mathrm{S}}\right], \\ x_1 &= \sin\left(\frac{\alpha}{2}\right)\left[\cos\left(\frac{\alpha}{2}\right)(\sin\theta_{\mathrm{E}}\cos\theta_{\mathrm{S}}\sin\phi_{\mathrm{E}} - \cos\theta_{\mathrm{E}}\sin\theta_{\mathrm{S}}\sin\phi_{\mathrm{S}}) + \sin\left(\frac{\alpha}{2}\right)\sin\theta_{\mathrm{E}}\cos\phi_{\mathrm{E}}\right], \\ y_1 &= \sin\left(\frac{\alpha}{2}\right)\left[\cos\left(\frac{\alpha}{2}\right)(\cos\theta_{\mathrm{E}}\sin\theta_{\mathrm{S}}\cos\phi_{\mathrm{S}} - \sin\theta_{\mathrm{E}}\cos\theta_{\mathrm{S}}\cos\phi_{\mathrm{E}}) + \sin\left(\frac{\alpha}{2}\right)\sin\theta_{\mathrm{E}}\sin\phi_{\mathrm{E}}\right], \\ z_1 &= \sin\left(\frac{\alpha}{2}\right)\left[\sin\left(\frac{\alpha}{2}\right)\cos\theta_{\mathrm{E}} - \cos\left(\frac{\alpha}{2}\right)\sin(\theta_{\mathrm{E}})\sin\theta_{\mathrm{S}}\sin(\phi_{\mathrm{S}}-\phi_{\mathrm{E}})\right]. \end{aligned}$$

It is easy to see that $\mathcal{F}_0 = \mathcal{F}_1 = \cos(\alpha/2)$. Plugging these expressions into Eq. (25) we get

$$f(\alpha,\theta_{\mathrm{S}},\theta_{\mathrm{E}},\phi_{\mathrm{S}},\phi_{\mathrm{E}})\cos\alpha + g(\alpha,\theta_{\mathrm{S}},\theta_{\mathrm{E}},\phi_{\mathrm{S}},\phi_{\mathrm{E}})^2 \geq 0, \tag{27}$$

where $g(\alpha,\theta_{\mathrm{S}},\theta_{\mathrm{E}},\phi_{\mathrm{S}},\phi_{\mathrm{E}}) = \boldsymbol{r}_0\cdot\boldsymbol{r}_1 - y_0 y_1$ and

$$f(\alpha,\theta_{\mathrm{S}},\theta_{\mathrm{E}},\phi_{\mathrm{S}},\phi_{\mathrm{E}}) = 1 - \cos^2\left(\frac{\alpha}{2}\right)\left\{\cos^2\theta_{\mathrm{E}} + \left[\sin\theta_{\mathrm{E}}\sin\theta_{\mathrm{S}}\cos(\phi_{\mathrm{S}}-\phi_{\mathrm{E}}) + \cos\theta_{\mathrm{E}}\cos\theta_{\mathrm{S}}\right]^2\right\}.$$

Consequently, the inequality (27) cannot be violated for $0 \leq \alpha \leq \pi/2$. ■

ECHO SEQUENCE AND DEPHASING

In this section we discuss the noises behind the classicalisation of our quantum memory we observe when introducing the waiting time.

First, recall that, in many laboratory environments, magnetic-field noise is not purely incoherent noise arising from an uncharacterised environment, but often it also contains *coherent* components at well-defined frequencies. Noise at the frequency of the mains electricity supply, and at its harmonics, is particularly difficult to control. In Europe, these components occur at 50 Hz and its multiples. Such noise cannot always be fully eliminated; instead, they can be addressed using active magnetic-field compensation. By doing so, we were able to suppress coherent noise at 50 Hz, but, unfortunately, we could not do the same for 100 Hz.

To address this issue, we mitigate the remaining 100 Hz component using dynamical decoupling [3]. Specifically, we apply a variable number of echo pulses after each 2.5 ms waiting interval. The resulting filter function minimises sensitivity to coherent noise at 100 Hz as well as to quasi-static (DC) noise. Although this procedure increases sensitivity at 50 Hz, this does not pose a problem because those components are already suppressed by the active field compensation.

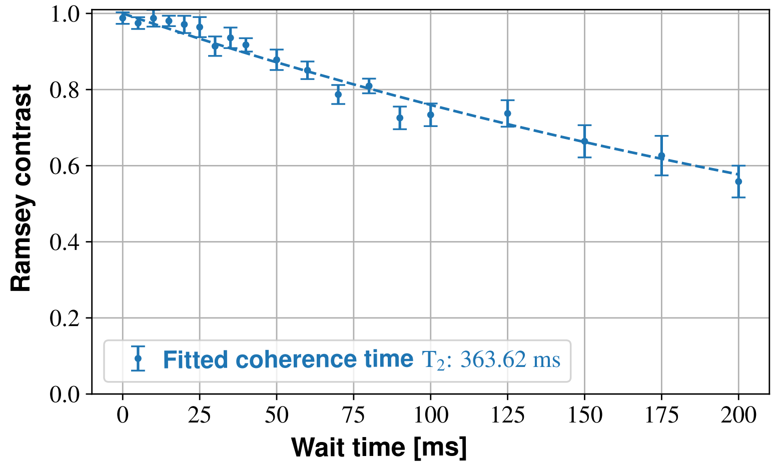

FIG. 1. **Coherence time measurement.** Measured dephasing time or effective coherence time T_2 of the trapped ion processor. Here, "Ramsey contrast" refers to the result of a standard Ramsey experiment. It informs how much coherence is left.

Finally, in order to simulate the expected decay rate, we measure the effective dephasing time of the dynamical decoupling sequence as well as the fidelity of the echo pulses used. Since always the exact same echo pulse is used in the gate sequence, calibration errors accumulate, yet we obtained an average fidelity of 0.995 per echo.

We measured the dephasing time with a set of Ramsey experiments (see e.g. Ref. [4]) over variable waiting time using an exponential fit. Of course, mathematically different models could be fitted to the data, yet the exponential fit is suitable for at least two reasons. First, the fit corresponds to frequency noise distributed like a Lorentzian around the centre frequency, which has a clear physical meaning and, second, as shown in Fig. 1, it fits the observed data with great precision (specially in comparison with other possible models like Gaussian). With this data at hand, we simulate the expected decay of our quantum memory in the main text. The agreement between our theory supplied with simple noise model and the observed decay shows the efficiency of our method to detect memory classicalisation.

ESTIMATING CROSS-TALK INFLUENCES

Finally, we discuss experimental inaccuracies that can lead to cross-talk influences. Since the absence of such causal influence is a central hypothesis which cannot be justified upon strong physical assumptions such as finite velocity of physical interactions (Einstein's relativity) which is often invoked in Bell's tests. In fact, as stressed in the main text, this assumption must be rigorously tested, which, as we have seen, can be done using ACDE and Pearl's inequality. We will therefore present the main physical reasons behind possible imperfections in our setup and provide some data that are used to estimate such detrimental causal influence.

In our experiment, various error channels connecting the two systems **A** and **E** can add new causal influences on the measurements undertaken, opening a loophole in our certification scheme. On a physical layer, the main cause for remaining crosstalk-influences is what is called crosstalk of the laser beams addressing the individual ions. Since the ions are only spaced several µm from

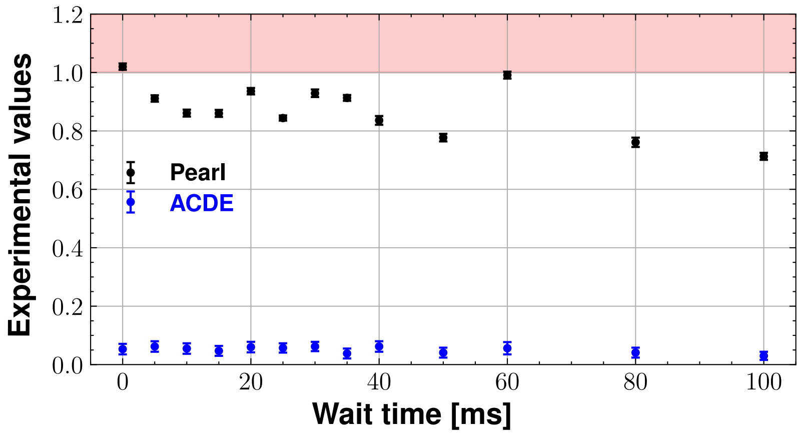

FIG. 2. **Pearl's inequality and the ACDE values over waiting time.** The threshold at which indicates cross-talk influences is given by the red-shaded area, corresponding to $I_{Pearl} > 1$. The ACDE-values can be used to test Eq. (**??**) from the main text, and are also found to present no problem for the certification of our quantum memory.

each other, laser beams have to be very tightly focussed to only address one ion at a time. Still, its finite size, limited by its wavelength, as well as imperfections in the beam shape (e.g. Airy disks) result in a residual intensity of light at the ions supposed to be left dark. The tilt with which we address the ion chain (67.5 °) amplifies this effect, since it increases the effective beam cross-section in the ion plain by nearly 10 %. Crosstalk is strongly mitigated by using a spacer ion in between the two implementing the protocol, pushing the logical error per gate below 10^{-3}. Still, we have to consider the effects it has on our certification scheme, whence the choice for "cross-talk influences" in our theoretical analysis.

At this point, some comments on device independence and associated loopholes are in order. First, as discussed in Section A, Pearl's inequality is formulated in terms of observational probabilities $\Pr(AB \mid X)$ and provides a *sufficient* condition for the presence of cross-talk influences; that is, if the inequality is not violated, the existence of crosstalk cannot be ruled out. The ACDE, by contrast, yields a direct estimate of cross-talk influences, but it relies on interventional probabilities $\Pr(B_{\mathrm{do}(A,X)})$.

In the specific implementation of the experiment (see Section E of this Supplementary Information), the same empirical probability table can be used to estimate both quantities. However, we emphasise that this does *not* close the corresponding loophole, since doing so breaks device-independence by attributing two distinct causal structures, related through intervention, to the same data set. Accordingly, the measured data should be understood as *addressing*, rather than *closing*, the aforementioned loophole.

For the quantum memory test, we estimated both Pearl's inequality and ACDE. The results with increasing waiting time are plotted in Fig. 2. For the case of an ideal quantum memory, the value we expect for Pearl's inequality to be

$$\frac{2+\sqrt{2}}{4} \approx 0.853. \tag{28}$$

This value is very close to the optimally measured, ≈ 0.883 [Eq. (8) in the main text]. Its behaviour while increasing the waiting time in shown in Fig. 2, where one can also find the corresponding values for ACDE.

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