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Quantum interference and correlation control of frequency-bin qubits: supplementary material

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This document provides supplementary information to "Quantum interference and correlation control of frequency-bin qubits," https://doi.org/10.1364/OPTICA.5.001455. We provide details on the experimental setup, theoretical predictions, and calculations of the visibility, entanglement witness, and density matrix.

1. EXPERIMENTAL DETAILS

We couple a continuous-wave Ti:sapphire laser (M Squared) into a fiber-pigtailed periodically poled lithium niobite (PPLN; SRICO) waveguide, temperature controlled at ∼85°C for spontaneous parametric down-conversion under type-0 phase matching. Spectrally entangled photon pairs spanning >2.5 THz are subsequently filtered by a Fabry-Perot etalon (Optoplex) with 25 GHz mode spacing (matched to the ITU grid and thus plug-in compatible with telecom fiber networks) to produce a biphoton frequency comb (BFC), with each comb line possessing a full-width at half-maximum linewidth of 1.8 GHz. The center frequency of the pump laser is carefully locked to align the generated signal-idler pairs with etalon peaks, i.e., to maximize coincidences between the spectrally filtered modes. We utilize a pulse shaper (BFC shaper; Finisar) to perform amplitude and phase filtering to prepare particular input states for quantum frequency processing.

Our quantum frequency processor (QFP) consists of two 40 Gb/s EOMs (EOSpace) with a pulse shaper [1] (QFP shaper; Finisar) sandwiched between them, with a total insertion loss of 12.5 dB [2]. We note that synchronizing the biphoton emission time to the EOM phase is not necessary when the two-photon coherence time is much larger than the modulation period. This condition is guaranteed, e.g., by a pump laser with narrow linewidth, as in the current experiments. To implement a Hadamard operation, i.e., frequency-bin beamsplitter, we drive the two EOMs with 25 GHz π-phase-shifted sinewaves, and apply a step function with π-phase jump between the two computational modes on the shaper. The specific phase patterns are obtained in advance from the optimization program in Refs. [2, 3], which achieve fidelity $\mathcal{F} = 0.9999$ and success probability $\mathcal{P} = 0.9760$ numerically for the Hadamard operation. (Experimentally, this system achieves near-perfect agreement with the numerical predictions [2].) The output photons are frequency-demultiplexed by an amplitude-only wavelength selective switch (WSS; Finisar) having 12.5 GHz channel specificity. Each time we route two different spectral modes (each takes up two pixels on the WSS) to two superconducting nanowire single-photon detectors (SNSPD; Quantum Opus) to record single counts as well as the coincidences within 1.5 ns bins.

2. QUANTUM OPERATIONS

The specific configuration for our Hadamard gate (cf. supplement of Ref. [2]) relies on the temporal phase modulation $\varphi(t) = \pm\Theta \sin \Delta\omega t$ ($\Theta = 0.8169$ rad) on the first and second EOMs, respectively. And for a gate operating on bins 0 and 1, the discrete pulse shaper phases can be written as

$$\phi_n = \begin{cases} \phi_0 & ; n \leq 0 \\ \phi_0 + \alpha & ; n \geq 1. \end{cases} \tag{S1}$$

Here ϕ_0 is an offset with no physical significance, while $\alpha = \pi$ for the ideal Hadamard. Yet α can be tuned as well; doing so actually permits tunable reflectivity. Specifically, if we write out the 2×2 transformation matrix on modes 0 and 1 as a function of this phase,

$$V = \begin{bmatrix} V_{00}(\alpha) & V_{01}(\alpha) \\ V_{10}(\alpha) & V_{11}(\alpha) \end{bmatrix}, \tag{S2}$$

we can define the variable reflectivities (i.e., mode-hopping probabilities) and transmissivities (probabilities of preserving frequency) as

$$\begin{aligned} \mathcal{R}_{0\to1} &= |V_{10}(\alpha)|^2 = \left|(1-e^{i\alpha})\sum_{k=1}^{\infty} J_k(\Theta)J_{k-1}(\Theta)\right|^2 \\ \mathcal{R}_{1\to0} &= |V_{01}(\alpha)|^2 = \left|(1-e^{i\alpha})\sum_{k=1}^{\infty} J_k(\Theta)J_{k-1}(\Theta)\right|^2 \\ \mathcal{T}_{0\to0} &= |V_{00}(\alpha)|^2 = \left|J_0^2(\Theta) + (1+e^{i\alpha})\frac{1-J_0^2(\Theta)}{2}\right|^2 \\ \mathcal{T}_{1\to1} &= |V_{11}(\alpha)|^2 = \left|e^{i\alpha}J_0^2(\Theta) + (1+e^{i\alpha})\frac{1-J_0^2(\Theta)}{2}\right|^2, \end{aligned} \tag{S3}$$

where $J_k(\Theta)$ is the Bessel function of the first kind. We note that, when $\alpha = \pi$, the elements $\{V_{00}, V_{01}, V_{10}\}$ are all real and positive, while V_{11} is real and negative—in accord with the ideal Hadamard and leading to destructive HOM interference between the reflect/reflect and transmit/transmit two-photon probability amplitudes. Additionally, these expressions satisfy $\mathcal{R}_{0\to1} = \mathcal{R}_{1\to0} \equiv \mathcal{R}$ and $\mathcal{T}_{0\to0} = \mathcal{T}_{1\to1} \equiv \mathcal{T}$. As α is tuned over $0 \to \pi \to 2\pi$, $\mathcal{R}$ follows from 0 to a peak of 0.4781 and back to 0, while $\mathcal{T}$ starts at 1, drops to 0.4979, and returns to 1. The sum $\mathcal{R} + \mathcal{T}$ defines the gate success probability, which drops slightly at $\alpha = \pi$ due to the use of single-frequency electro-optic modulation. These particular values are confirmed experimentally in Fig. 2(a) of the main text with coherent state measurements [2].

3. HONG-OU-MANDEL INTERFERENCE

The generated biphoton frequency comb can be described as a state of the form

$$|\Psi\rangle = \sum_{n=1}^{N} c_n |1_{\omega_{1-n}}\rangle_A |1_{\omega_n}\rangle_B, \tag{S4}$$

or in terms of bosonic mode operators,

$$|\Psi\rangle = \sum_{n=1}^{N} c_n \hat{a}_{1-n}^\dagger \hat{a}_n^\dagger |\text{vac}\rangle_A |\text{vac}\rangle_B, \tag{S5}$$

where $\hat{a}_n$ ($\hat{a}_n^\dagger$) annihilates (creates) one photon in the frequency bin centered at ω_n, and the coefficients c_n are set by a pulse shaper [BFC Shaper in Fig. 2(a)]. The A and B nomenclature defines the modes held by each of two parties: A consists all ω_n such that $n \leq 0$, B everything with $n \geq 1$. We favor this notation over the more traditional "signal" and "idler" classification because (i) our frequency operations can move photons between A and B mode sets—and indeed *does* in the case of HOM—and (ii) there are no other distinguishing degrees of freedom to label the photons.

Our quantum frequency processor transforms these bins into outputs $\hat{b}_m$ (at frequencies ω_m) via

$$\hat{b}_m = \sum_{n=-\infty}^{\infty} V_{mn}\hat{a}_n. \tag{S6}$$

The matrix V describes the entire operation over all modes. Then at the output we measure the spectrally resolved coincidences between bins n_A and n_B, i.e.,

$$C_{n_A n_B} = \langle\Psi|\hat{b}_{n_A}^\dagger \hat{b}_{n_B}^\dagger \hat{b}_{n_B}\hat{b}_{n_A}|\Psi\rangle, \tag{S7}$$

as well as the singles

$$S_n = \langle\Psi|\hat{b}_n^\dagger \hat{b}_n|\Psi\rangle. \tag{S8}$$

In the case of HOM interference, we filter out all photon pairs except c_1 [Eq. (S4)], so the input state is $|\Psi\rangle = |1_{\omega_0}\rangle_A|1_{\omega_1}\rangle_B$, which gives $C_{01} = |V_{00}V_{11} + V_{01}V_{10}|^2$ and $S_n = |V_{n0}|^2 + |V_{n1}|^2$. In light of the previous discussion on beamsplitter tunability, we thus predict:

$$\begin{aligned} C_{01} &= |\mathcal{R}(\alpha) - \mathcal{T}(\alpha)|^2 \\ S_0 = S_1 &= \mathcal{R}(\alpha) + \mathcal{T}(\alpha) \\ S_{-1} = S_2 &\approx 1 - \mathcal{R}(\alpha) - \mathcal{T}(\alpha), \end{aligned} \tag{S9}$$

where the nonunity success probability [$\mathcal{R}(\pi) + \mathcal{T}(\pi) = 0.976$] results in some photons scattering into bins -1 and 2. (Scattering beyond these modes is not observable in experiment, consistent with the theoretical prediction of only $\sim 10^{-4}$ probability to leave the center four bins.) Invoking the theoretically predicted values for $\mathcal{R}$ and $\mathcal{T}$, we use weighted least-squares to fit the function $f(\alpha) = K_0 + K_1 C_{01}(\alpha)$ to the data in Fig. 3(b) and extract the visibility

$$\mathcal{V} = 1 - \frac{f(\pi)}{f(0)} = \frac{K_1[C_{01}(0) - C_{01}(\pi)]}{K_0 + K_1 C_{01}(0)} = 0.971 \pm 0.007. \tag{S10}$$

Now, because the singles S_0 and S_1 drop slightly at $\alpha = \pi$ [cf. Fig. 3(b) of the main text]—which is not the case in a traditional HOM experiment—we also look at the visibility of the normalized cross-correlation function, $g_{01}^{(2)} = \frac{C_{01}}{S_0 S_1}$. For in the most pathological case, a reduction in the unnormalized coincidences C_{01} could in principle be due to dropping singles S_0 or S_1, which would not be surprising from a classical view: if one detector rarely clicks, of course its coincidences with another detector will drop as well. On the other hand, the normalized $g_{01}^{(2)}$ does not suffer from this issue, by accounting for singles counts directly. Accordingly, we repeat the least-squares fit using the theoretically predicted $g_{01}^{(2)}(\alpha)$, along with the measured coincidences [Fig. 3(b)] divided by the product of mode 0 and 1 single counts [Fig. 3(c)]. In this more conservative case, we still retrieve $\mathcal{V} = 0.967 \pm 0.007$, fully confirming the nonclassicality of our HOM interference.

4. QUANTUM STATE MANIPULATION

For the state rotation experiments, we filter out all modes except four, leaving the entangled qubits [$n = 4, 5$ in Eq. (S4)]:

$$|\Psi\rangle = \frac{1}{\sqrt{2}}\left(|1_{\omega_{-3}}\rangle_A|1_{\omega_4}\rangle_B + |1_{\omega_{-4}}\rangle_A|1_{\omega_5}\rangle_B\right). \tag{S11}$$

Ideally, parametric downconversion and filtering should produce this relative phase automatically; but in order to compensate any residual dispersion prior to the quantum gates, we also fine-tune the phase with the BFC pulse shaper, experimentally maximizing spectral correlations in the $H_A \otimes H_B$ measurement case (see below). We have six initially empty modes between those populated in A and B, allowing us to apply combinations of Hadamard operations and the identity to each pair of modes—$\{-4,-3\}$ and $\{4,5\}$—without any fear of the photon in A jumping over to B's modes, and vice versa (cf. guardband discussion in Ref. [2]). Accordingly, after the frequency-bin transformation V (chosen to apply the desired joint operation), the coincidence probability for any $(n_A \leq 0, n_B \geq 1)$ is given by

$$C_{n_A n_B} = |V_{n_A,-3}V_{n_B,4} + V_{n_A,-4}V_{n_B,5}|^2 . \tag{S12}$$

This expression accounts for all aspects of the potentially nonideal mode transformation. Focusing on the qubit modes ($n_A \in \{-4,3\}, n_B \in \{4,5\}$), we have the ideal coincidences under all four cases of Fig. 4 as:

$$\begin{aligned}
V(\mathbb{1}_A \otimes \mathbb{1}_B) \Longrightarrow \quad C_{n_A n_B}^{\mathbb{1}_A \otimes \mathbb{1}_B} &= \frac{1}{2}\left(\delta_{n_A,-3}\delta_{n_B,4} + \delta_{n_A,-4}\delta_{n_B,5}\right) \\
V(\mathbb{1}_A \otimes H_B) \Longrightarrow \quad C_{n_A n_B}^{\mathbb{1}_A \otimes H_B} &= \frac{1}{4}\left(\delta_{n_A,-3}\delta_{n_B,4} + \delta_{n_A,-3}\delta_{n_B,5} + \delta_{n_A,-4}\delta_{n_B,4} + \delta_{n_A,-4}\delta_{n_B,5}\right) \\
V(H_A \otimes \mathbb{1}_B) \Longrightarrow \quad C_{n_A n_B}^{H_A \otimes \mathbb{1}_B} &= \frac{1}{4}\left(\delta_{n_A,-3}\delta_{n_B,4} + \delta_{n_A,-3}\delta_{n_B,5} + \delta_{n_A,-4}\delta_{n_B,4} + \delta_{n_A,-4}\delta_{n_B,5}\right) \\
V(H_A \otimes H_B) \Longrightarrow \quad C_{n_A n_B}^{H_A \otimes H_B} &= \frac{1}{2}\left(\delta_{n_A,-4}\delta_{n_B,4} + \delta_{n_A,-3}\delta_{n_B,5}\right),
\end{aligned} \tag{S13}$$

where $\delta_{nm} = \{1 \text{ if } n = m, 0 \text{ if } n \neq m\}$. These expressions predict perfect negative correlations for the case $\mathbb{1}_A \otimes \mathbb{1}_B$—i.e., detecting the low frequency of A occurs in coincidence with the high frequency of B, and vice versa—while positive correlations result for $H_A \otimes H_B$. For the other two cases, no frequency correlations are present, with all four combinations equally likely.

The predictions of Eq. (S13) are precisely those of the EPR paradox [4], particularly the discrete version formulated by Bohm [5]. Applying unitaries $\mathbb{1}$ and H followed by frequency detection produces measurements of the Pauli Z and X bases, respectively, so that our simultaneous correlations in two mutually unbiased bases reveals the strange characteristics of quantum mechanics, although admittedly without verifying, in our experiments, its nonlocal character. Finally, while we formulate the theory here viewing the operations as *measurements*—as this is most direct in explaining our results—they can equivalently be considered as *gates* on the input state, a useful distinction in considering more complex frequency networks where the photons are processed further before detection.

5. ENTANGLEMENT WITNESS

The EPR paradox exemplified by the theory of Eq. (S13) and results in Fig. 4 can be quantified in terms of an entropic entanglement witness between party A and B. Adapting Eq. (329) of Ref. [7] to our experiments gives the following inequality, satisfied by all separable states:

$$\mathcal{H}(\mathbb{1}_A|\mathbb{1}_B) + \mathcal{H}(H_A|H_B) \geq q_{MU}, \tag{S14}$$

where $\mathcal{H}(U_A|U'_B)$ is the entropy of A's detected frequency bin, given knowledge of B's result, when unitary operations U and U' are applied. (We use the transformations as arguments, rather than the observables per se, for maximal clarity with our experimental configuration.) The Maassen-Uffink bound q_{MU} [6, 7] depends on the overlap between the frequency basis vectors in the rows of $\mathbb{1}_{\mathbb{A}}$ and H_A, which can be written in terms of the beamsplitter reflectivity $\mathcal{R}$ and transmissivity $\mathcal{T}$ (defined above) as

$$q_{MU} = -\log_2 \max\left(\left\{\frac{\mathcal{R}}{\mathcal{R}+\mathcal{T}}, \frac{\mathcal{T}}{\mathcal{R}+\mathcal{T}}\right\}\right), \tag{S15}$$

where $\{\mathcal{R},\mathcal{T}\}$ are evaluated at $\alpha = \pi$. Because we postselect on coincidences in the two-qubit Hilbert space, we have normalized $\{\mathcal{R},\mathcal{T}\}$ by the success probability; plugging in theoretical values, we obtain $q_{MU} = 0.9710$—close to the maximum of 1 for perfect mutually unbiased bases in $d = 2$ dimensions ($\log_2 d$).

To calculate the conditional entropies corresponding to the measurements in Fig. 4, we employ Bayesian mean estimation (BME) on the raw count data [8, 9]. Unlike alternative approaches, such as maximum likelihood estimation, BME yields error bars directly for any computed function and incorporates prior knowledge into the calculation naturally. To produce as conservative an estimate as possible, we make no specifying assumptions about the underlying state. For each situation in Fig. 4, we posit a three-parameter multinomial likelihood function (four probabilities minus normalization), with counts taken directly from the raw data; we take the prior as uniform. The estimated means and standard deviations of the conditional entropies are then

$$\begin{aligned}
\mathcal{H}(\mathbb{1}_A|\mathbb{1}_B) &= 0.19 \pm 0.03 \\
\mathcal{H}(H_A|\mathbb{1}_B) &= 0.997 \pm 0.003 \\
\mathcal{H}(\mathbb{1}_A|H_B) &= 0.993 \pm 0.005 \\
\mathcal{H}(H_A|H_B) &= 0.29 \pm 0.04.
\end{aligned} \tag{S16}$$

As expected, the mismatched bases have near-maximal entropy (1 bit), while matched cases are much lower. We emphasize that the full effect of accidentals are included in these numbers; appreciably lower matched entropies may be possible in a model incorporating dark counts as well. Nonetheless, summing these entropies directly gives $\mathcal{H}(\mathbb{1}_A|\mathbb{1}_B) + \mathcal{H}(H_A|H_B) = 0.48 \pm 0.05$—violating the bound q_{MU} by 9.8 standard deviations, and thereby confirming the nonseparability of our quantum state with high confidence.

6. STATE RECONSTRUCTION

To estimate the complete two-qubit density matrix, we again employ BME [8, 9] but now with the assumption of a single quantum state underlying all four measurements in Fig. 4. As noted above, these four combinations are equivalent to joint measurements of the two-qubit observables $\{Z_A \otimes Z_B, X_A \otimes Z_B, Z_A \otimes X_B, X_A \otimes X_B\}$, where the identity $\mathbb{1}$ permits measurement of Z, and the unitary H allows measurement of X. Despite the fact this set of measurements is tomographically incomplete, we are nevertheless able to infer a complete state estimate, with appropriately higher uncertainties in the unmeasured bases (e.g., Pauli Y). Finally, we emphasize that experimentally we only have access to the detector click (or no-click) events that are more naturally described in terms of positive-operator valued measures (POVMs) rather than von Neumann type projectors on the eigenvectors of Pauli X and Z operators.

For a specific two-qubit observable and chosen pair of frequency bins, we have the POVMs $\Lambda^{(A)} = \{\hat{\Pi}^{(A)}, \mathbb{1} - \hat{\Pi}^{(A)}\}$ for subsystem A, and $\Lambda^{(B)} = \{\hat{\Pi}^{(B)}, \mathbb{1} - \hat{\Pi}^{(B)}\}$ for subsystem B, where $\hat{\Pi}^{(A,B)}$ correspond to photon clicks, $\mathbb{1} - \hat{\Pi}^{(A,B)}$ to the absence of a click. Absence of a click can be due to detection inefficiency or the photon being in an unmonitored mode. An outcome of a two-qubit POVM $\Lambda^{(A)} \otimes \Lambda^{(B)}$ will fall into one of the three experimentally recorded numbers: coincidence counts (C_{AB}), singles counts on detector A (S_A), and singles counts on detector B (S_B). These form our specific data set $\mathcal{D} = \{C_{AB}, S_A, S_B\}$. In our model, we assume fixed channel efficiencies for A and B propagation and detection (η_A and η_B), and the following normalized probabilities under no loss and perfect detection: p_{AB} (coincidence, one photon in mode A and one photon in mode B), p_{A0} (one photon in mode A and no photon in mode B), p_{0B} (one photon in mode B and no photon in mode A), p_{00} (no photon in mode A or B).

Letting N denote the number of photon pairs generated in the measured time interval, we can enumerate the following four experimental possibilities, formed by the products of all operators from this POVM pair. (i) $\hat{\Pi}^{(A)} \otimes \hat{\Pi}^{(B)}$: coincidence between detectors A and B. This occurs with probability $\eta_A \eta_B p_{AB}$ and is observed C_{AB} times. (ii) $\hat{\Pi}^{(A)} \otimes [\mathbb{1} - \hat{\Pi}^{(B)}]$: click on detector A, no click on B. This occurs with probability $\eta_A [p_{AB}(1-\eta_B) + p_{A0}]$ and is observed $S_A - C_{AB}$ times. (iii) $[\mathbb{1} - \hat{\Pi}^{(A)}] \otimes \hat{\Pi}^{(B)}$: no click on detector A, click on detector B. This has probability $\eta_B [p_{AB}(1-\eta_A) + p_{0B}]$ and is observed $S_B - C_{AB}$ times. (iv) $[\mathbb{1} - \hat{\Pi}^{(A)}] \otimes [\mathbb{1} - \hat{\Pi}^{(B)}]$: no click on either detector. This occurs with probability $p_{AB}(1-\eta_A)(1-\eta_B) + p_{A0}(1-\eta_A) + p_{0B}(1-\eta_B) + p_{00}$ and is counted $N - S_A - S_B + C_{AB}$ times. Our likelihood function, $P(\mathcal{D}|\beta)$, is then a multinomial distribution over the aforementioned probabilities and outcomes, where $\beta = \{\hat{\rho}, \eta_A, \eta_B, N\}$ is the underlying parameter set. The idealized probabilities $\{p_{AB}, p_{A0}, p_{0B}, p_{00}\}$ are all functions of the density matrix $\hat{\rho}$, which we limit to physically allowable states [9].

Up to this point, we have focused on a *specific* choice of POVMs, $\Lambda^{(A)} \otimes \Lambda^{(B)}$. To account for all 16 POVM combinations (basis pairs and frequency-bin pairs) in the two-qubit space of Fig. 4, we form the product over all settings, leaving the complete posterior distribution

$$P(\beta|\boldsymbol{\mathcal{D}}) = \frac{\left[\prod_j P(\mathcal{D}_j|\beta)\right] P(\beta)}{P(\boldsymbol{\mathcal{D}})}, \tag{S17}$$

where the bolded $\boldsymbol{\mathcal{D}}$ represents the union of the respective results $\mathcal{D}_j$ from each particular setting ($j = 1, 2, ..., 16$). Our prior $P(\beta)$ is taken to be uniform in a Haar-invariant sense, and the marginal $P(\boldsymbol{\mathcal{D}})$ is found by integrating the numerator in Eq. (S17). With this posterior distribution, we can estimate any parameter of interest via integration, such as the mean density matrix

$$\hat{\rho}_{\mathrm{BME}} = \int d\beta \; P(\beta|\boldsymbol{\mathcal{D}})\hat{\rho}. \tag{S18}$$

Due to the complexity of integrals of this form, we employ numerical slice sampling for their evaluation [10]. The resulting estimates are discussed in the main text and plotted in Fig. 5.

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