

Hong-Ou-Mandel two-photon x-ray states

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We have observed Hong-Ou-Mandel interference of high-brightness synchrotron x-rays with a Mach-Zehnder interferometer, yielding potential two-photon states of interest for x-ray quantum optics.

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Entanglement is a key aspect in the theoretical description of many quantum materials, but experimental techniques for characterizing entanglement are quite limited. The use of entangled probes to detect entanglement has been suggested and is being pursued with neutrons, for example [1]. X-rays are powerful probes of material properties, and there are promising efforts to create entangled multiphoton x-ray states that could become useful quantum probes [2,3]. We report here on a new approach to creating multiphoton x-ray states utilizing a beam splitter, the Hong-Ou-Mandel (HOM) effect [4], at a high-brightness synchrotron x-ray source.

Two indistinguishable photons incident on opposite entrance ports of an ideal beam splitter yield the output state

$$|\psi\rangle_{11} = (t^2 - r^2)|11\rangle + \sqrt{2}tr(|20\rangle - |02\rangle), \quad (1)$$

where the subscript 11 refers to the two input photons, r and t are the real reflection and transmission coefficients, and $|nm\rangle$ denotes the number of photons in each of the two exit ports (see Appendix A of Ref. [5]). Note that intensity interference causes the first term to go to zero for a 50-50 beam splitter.

The second term is the entanglement of the two-photon exit configurations, a N00N state ($N = 2$) [6], and is the basis for our approach to multiphoton x-ray states. HOM utilized pairs of photons generated by parametric down-conversion (PDC) in a pure number state. Our source is an x-ray synchrotron where the photon beam can be modeled as a mixture of number states, where each number state $|\psi_\alpha\rangle$ is present with a probability p_α . The experiment optimizes the probability that a $|11\rangle$ state reaches the beam splitter and compares the postselected output to Eq. (1).

We report two experimental results using x-ray synchrotron radiation (Fig. 1). First, we observe an $\sim 2\%$ HOM dip in the $|11\rangle$ output with a corresponding bump in the two-photon yield. Second, we find a larger width than would be allowed for photons arriving simultaneously at the beam splitter. We discuss these below, after a description of the experiment.

The x-ray source is a monochromated 8 keV undulator beam at the Advanced Photon Source (APS), a high-brightness synchrotron [7]. Circulating 6 GeV electron bunches with a pulse duration of ~ 230 ps (FWHM) generate x-rays as they pass through the ~ 200 magnetic poles of an undulator. The electromagnetic wave amplitudes from any individual electron constructively interfere to produce the 8 keV x-rays near the peak of the undulator spectrum. Wave amplitudes from different electrons do not interfere [8]. Thus, this source consists of the incoherent sum of x-rays from independent electrons in each bunch. The resulting photon statistics correspond to chaotic light, as confirmed, for example, by intensity interferometry measurements of the degree of second-order coherence at a similar source [9].

Experimental details. Two-photon interference with a synchrotron source first requires the generation of indistinguishable photons, then pairs of these are split into separate paths, redirected toward a scanning beam splitter that samples the region where both beams can overlap, and finally a multiphoton detection system counts the number of photons in each exit beam for each synchrotron pulse. The need to split and recombine the beams naturally calls for a Mach-Zehnder interferometer design [Fig. 1(b)].

The synchrotron beam is set to the energy $E = 8$ keV with bandwidth $\Delta E = 74$ meV utilizing upstream beamline monochromators; see the Supplemental Material for details of the synchrotron optics and the HOM apparatus [10] (see also Ref. [11] therein). The specially constructed Mach-Zehnder interferometer had a single monolith for the two Bragg x-ray mirrors and thinned silicon crystals for the two beam splitters, all oriented for Si(400) diffraction in the Bragg geometry [12].

Both splitters are controlled by precision linear and rotary stages, critical for ensuring that all elements are diffracting optimally and the beam paths intersect precisely at the second beam splitter. Following HOM, the beam splitter is translated in small increments Δx to vary its overlap with the two beams. Slits were reduced until on average only one photon was in each beam striking the final beam splitter for each synchrotron pulse, a five-order-of-magnitude reduction from the full undulator beam intensity. A typical beam size was $100 \mu\text{m} \times 100 \mu\text{m}$, less than the $140 \mu\text{m}$ lateral coherence length of the source.

Measurements require counting the integer number of photons at each detector for every synchrotron bunch. The

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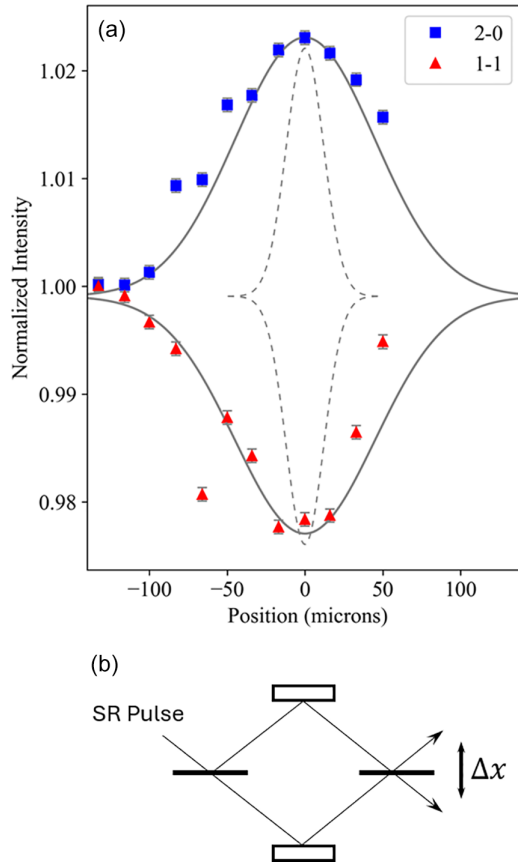


FIG. 1. HOM x-ray interference. (a) The (1, 1) and the (2, 0) and (0, 2) rates are plotted vs beam splitter position Δx , revealing the HOM dip and peak. The data are fit by a Gaussian $\exp[-\Delta x^2/l_c^2]$, where l_c is the lateral coherence length as defined by upstream slits [13]. The best fit shown used an FWHM of 106 μm , in good agreement with the 100 μm slit setting. Dashed lines are predicted curves assuming l_c is the 8 μm longitudinal coherence length for simultaneous photons. (b) Mach-Zehnder interferometer geometry with Bragg beam splitters and mirrors, with a synchrotron pulse input.

voltage outputs of photon-number-resolving avalanche photodiode detectors are digitized and analyzed before the next pulse arrives [14]. This requires the independent analysis of 13 million discrete events per second (i.e., the bunch frequency), a new experimental modality where a distinct measurement is recorded for each bunch. Output signals are converted to an integer number of photons based on prior calibration, so that one bunch yields one pair of integers.

Results. The experiment simply requires counting the number of (1, 1), (2, 0), and (0, 2) pairs as the beam splitter is translated. Figure 1 shows the dip in the normalized (1, 1) coincidence counts versus Δx . Also plotted is the corresponding peak in the combined (2, 0) and (0, 2) counts, clearly showing that the (1, 1) output is being converted into (2, 0) and (0, 2) outputs. We argue below that these excess (2, 0) and (0, 2) counts are actually the two-photon states in Eq. (1), since a classical explanation can be ruled out. Each data point is the sum of three scans each of 2 s duration, with error bars determined from counting statistics. Larger fluctuations are

caused by transient variations in the diffracting elements (see the Supplemental Material for more details [10]). With the incident flux set to one photon per beam per bunch, this dip corresponds to the interference of $\sim 3 \times 10^4$ photon pairs per second.

Why is there only a 2% dip? Because often the two photons are not truly indistinguishable. Indistinguishable photons from a synchrotron source must occupy the same source mode; photons from different modes in principle are distinguishable. Having beam slits smaller than the lateral coherence length ensures that all photons share the same lateral mode coordinates, but there are many possible longitudinal mode coordinates because the photon coherence time is much shorter than the pulse duration (28 fs versus 230 ps FWHM). If a single mode is occupied by two photons and one of them enters the interferometer, however, the conditional probability of the second doing the same is essentially unity (absent air absorption, monochromator distortions, etc.). Hence, the size of the HOM dip will be determined by the probability of modes being occupied by two photons, not the total number of possible modes.

The mean number of photons per mode is known as the photon degeneracy factor n_{ph} , which depends on the peak beam brightness, $n_{\text{ph}} = B_{\text{peak}} \lambda^3 / 4\sqrt{2} c$, where λ is the wavelength and c is the speed of light [15]. Assuming a Poisson distribution, we show below that having a 2% HOM dip corresponds to a photon degeneracy of $n_{\text{ph}} \sim 0.3$. This is within the range expected using the nominal brightness of the APS under the actual running conditions for these measurements; an independent measurement of B_{peak} is not available. Note also that having two photons in the same mode does not mean that they were emitted by the same electron or at the same time. Two electrons might be widely separated along the bunch but execute the same path through the undulator such that their emitted x-rays can be in the same mode but separated in time.

HOM found that interference goes away when the beam splitter was displaced by more than the photon's longitudinal coherence length. This response is indicated by the dashed curves in Fig. 1. Why is the actual dip so much broader? Because the two interfering x-rays do not arrive at the same time. A sketch of the real-space overlap of the two beams at the final beam splitter is shown in Fig. 2. Indistinguishability requires that the two exit photons must originate from the same region on the beam splitter, which is possible only inside the region outlined by the large diamond (height ΔX). If the two photons arrive simultaneously, however, the overlap of their longitudinal coherence lengths ($\sim 8 \mu\text{m}$) is limited by the smaller diamond of height Δx . Comparison with the data reveals that the observed width is governed by the spatial overlap region (large diamond) and not by the overlap of simultaneous photons (small diamond).

For HOM, the photon pairs were created in the same instant, arrived simultaneously at the beam splitter, and reached the detectors at the same time. Later, it was shown that the photons do not have to be born at the same time or even from the same source [16–18], and need not arrive at the beam splitter or the detectors simultaneously [19–21]. What is necessary is that the exit trajectories when both photons are transmitted (tt) and when both are reflected (rr) are indistinguishable.

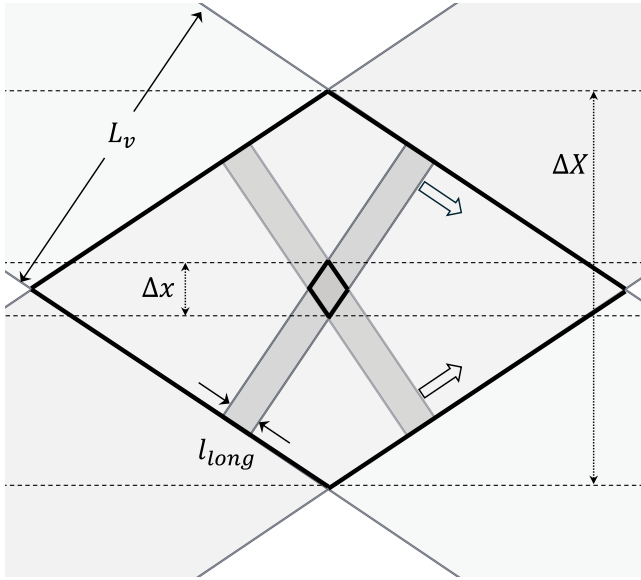


FIG. 2. Schematic of x-ray beam overlap. The large diamond maps out the region where the two incident beams overlap in space; the beam width L_v is set by the upstream vertical slit. A beam width of $100\ \mu\text{m}$ corresponds to a geometric height ΔX of $\sim 120\ \mu\text{m}$. The smaller diamond is the instantaneous region of overlap of two simultaneous x-rays, as defined by the $8\ \mu\text{m}$ longitudinal coherence length. Δx is the beam splitter range that samples temporally overlapping photons, with a geometric height of $\sim 14\ \mu\text{m}$. ΔX spans the entire overlap region including where the photons cannot be simultaneously present, which closely matches the region where interference was observed.

Then, the probability amplitudes for tt and rr cancel [see Eq. (1)], leaving only the entangled doubly occupied tr and rt exit beam states.

Figure 3 illustrates this process for the Mach-Zehnder interferometer. For a given electron bunch, consider two photons in the same mode and separated by a time interval Δt . When the two photons take different paths, the lead photon could take the lower path (Case A) or the upper path (Case B). Looking only at the exit trajectories after the final beam splitter, it is seen that rr_A is indistinguishable from tt_B , as are rr_B and tt_A . That is, not knowing which path is taken creates the HOM interference.

Discussion. We have explained how the lack of indistinguishability is responsible for a dip of only $\sim 2\%$, and that the Mach-Zehnder geometry allows for two photons separated in time to produce the observed interference width. The remaining question is: Does the two-photon bump in the data really correspond to entangled N00N states? Proof would require an x-ray version of the standard Clauser-Horne-Shimony-Holt (CHSH) Bell test [22], but we address two related questions below: Can classical fields explain these results, and is it even possible for a synchrotron source to exhibit observable quantum x-ray photon behavior?

The interaction of classical waves can produce some of the same interference phenomena observed with quantum states, so it is important to consider a classical explanation of our results. Both classical and quantum sources can be characterized by the degree of second-order coherence, $g^{(2)}$.

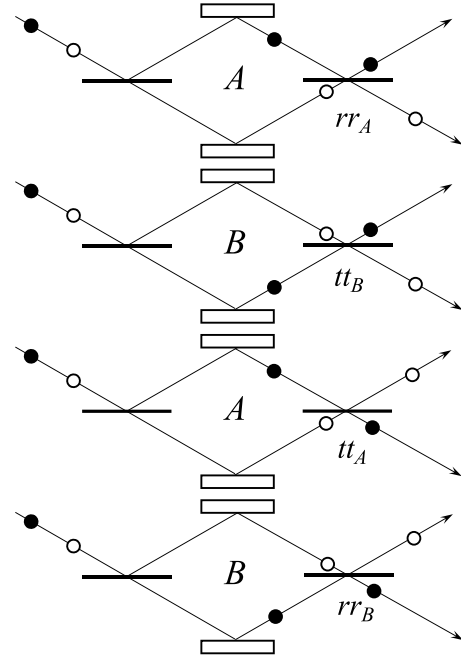


FIG. 3. Indistinguishable output trajectories for time-separated pairs. Two separated photons (filled and empty circles) enter the Mach-Zehnder interferometer and can take different paths. Case A is when the leading photon takes the lower path, and for Case B it takes the upper path. These are also shown approaching the HOM beam splitter, and again on their exit paths toward the detectors. Note that Case A with both reflected (rr_A) is indistinguishable from Case B with both transmitted (tt_B), and the same for tt_A and rr_B .

In general, the allowed classical range is $1 \leq g^{(2)} \leq \infty$, while the quantum range is $0 \leq g^{(2)} \leq \infty$ [23]. Measurements at x-ray synchrotrons yield $g^{(2)} \approx 1$ [9,24], so this does not by itself distinguish between classical and quantum. Classical interference, however, depends on the cross-correlation of electric fields for the two pulses where the interference terms contain $E_1^*(t)E_2(t - \Delta t)$ [25]. If one pulse has a finite amplitude and the other is time delayed by more than the coherence time, the second pulse will have practically zero amplitude and there can be no classical interference. This holds true here because the data cannot be explained by the photons arriving within the coherence time.

We can now address whether quantum phenomena can be observed with a synchrotron source. There is already published evidence for observing quantum interference from mixtures of number states [26]. Perhaps, the most remarkable is the HOM interference observed when one photon in a number state (from a quantum dot) is combined with light from the Sun, the epitome of a thermal source [27]. Not only was a large HOM dip seen, consistent with the first term in Eq. (1), two-photon entanglement was also established, confirming the second term in Eq. (1). We are taking this just one step further, replacing the quantum dot by a second electron from the same synchrotron bunch. We therefore conclude that quantum interference of number states from the mixed-state source is responsible for two-photon states as seen in the (2, 0) and (0, 2) bump in the data, while noting again that proof of entanglement will require an x-ray CHSH test.

To understand how the HOM dip depends on the photon degeneracy factor, let us first consider pure number states incident on the Mach-Zehnder interferometer, again using results derived from Ref. [5]. Because we postselect for when the number of detected photons is 2, we only consider the incident state $|20\rangle$, i.e., two indistinguishable photons at one entrance port and zero at the other. Then, the output state from the first beam splitter is

$$|\psi\rangle_{20} = t^2|20\rangle + r^2|02\rangle - \sqrt{2}tr|11\rangle. \quad (2)$$

Hence, the input to the second beam splitter contains $|20\rangle$, $|02\rangle$, and $|11\rangle$ terms. The final beam splitter output from $|11\rangle$ is given by Eq. (1), and the output from $|20\rangle$ is again given by Eq. (2). The $|02\rangle$ output is

$$|\psi\rangle_{02} = t^2|02\rangle + r^2|20\rangle + \sqrt{2}tr|11\rangle. \quad (3)$$

Note that the $|11\rangle$ input state will produce no $|11\rangle$ output states (coincident pairs) when $r = t$ [Eq. (1)], but the $|20\rangle$ and $|02\rangle$ will. Using Kim *et al.*'s derivation of the output of a Mach-Zehnder interferometer for arbitrary input number states when $r = t$ [see Eq. (28) of Ref. [28]], we find the expectation value for the $|11\rangle$ output coincidence rate at the exit is

$$\langle 1, 1 \rangle = 1 - \frac{1}{2} \overline{\sin^2 \theta} - \overline{\cos^2 \theta} = \frac{1}{4}, \quad (4)$$

where the last step averages over phase shifts θ between the interferometer paths. This is necessary because the beam splitter surface quality is insufficient for amplitude interference at x-ray wavelengths. Note that while the $|11\rangle$ state incident on a single beam splitter yields a 100% HOM dip [Eq. (1)], 75% is the maximum possible dip for a $|20\rangle$ input at a Mach-Zehnder interferometer. This means that the $\sim 2\%$ dip observed in the data could not be larger than 75% even if there were only a single mode and all incident photons were in pure number states.

Recall that we model the x-ray beam as a mixture of number states. Experimentally, we find that the exit beam distributions at the two detectors are quite accurately governed by Poisson statistics; this is also consistent with chaotic photon statistics when the detector time resolution exceeds the photon coherence time (see Chap. 6 of Ref. [23]). The probability for a given number state $|n\rangle$ in a single beam then is $p_n = k^n \exp(-k)/n!$, where k is the mean number of photons per bunch.

The Poisson distribution gives the probability of $|11\rangle$ incident on the final beam splitter as $p_1 p_1 = k^2 e^{-2k}$, while for $|20\rangle$ and $|02\rangle$ it is $p_2 p_0 = p_0 p_2 = k^2 e^{-2k}/2 = p_1 p_1/2$. In the wings where there is no interference, all three of these inputs have a 50% probability of producing a coincident pair output state, but half of the total originates from the $|11\rangle$ input. When the beam splitter is centered, the coincident rate from $|11\rangle$ goes to zero, while the others are unaffected. Hence, the maximum HOM dip from this mixed state is 50% (as opposed to the 75% dip for number states). For a given n_{ph} , the HOM dip will be half the probability of two photons occupying a single mode, $0.5(n_{\text{ph}}^2 \exp(-n_{\text{ph}})/2)$. This is plotted versus n_{ph} in Fig. 4, providing a definite prediction for enhancing the HOM dip and the two-photon state yield by increasing

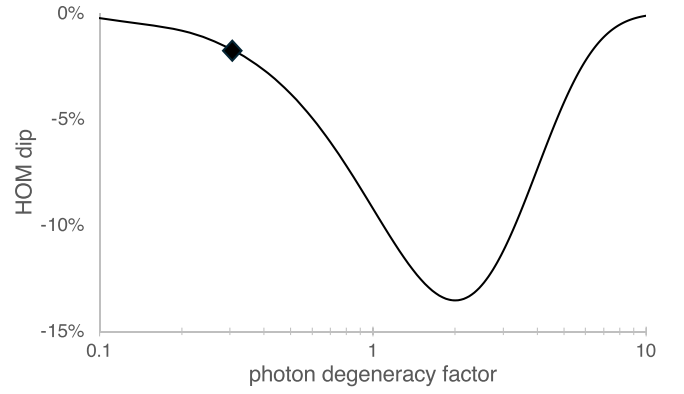


FIG. 4. Predicted HOM dip vs photon degeneracy factor. Symbol (◆) indicates current measurement.

the photon degeneracy factor. While the two-photon HOM dip declines at higher values of n_{ph} , higher-order quantum interference effects will then become possible.

Advances in x-ray sources have led to a growing interest in multiphoton diffraction processes. This includes experimental progress at an x-ray free electron laser (XFEL) [29] and theoretical considerations of stimulated multiphoton diffraction at high intensities [15,30]. This complements extensive progress made with PDC-generated x-rays [2,3] in recent years. The two-photon x-ray states created by HOM interference offer a new type of multiphoton state. It requires high brightness to achieve sufficient photon degeneracy, but not the high instantaneous intensities of an XFEL. The yields are substantially greater than what is currently achievable with x-ray PDC, but background issues are more challenging. The potential areas of application are likely to be different from those with XFEL and PDC multiphotons. Future efforts would include increasing the HOM dip with improved x-ray optics and higher brightness, exploring the unique absorption properties of two-photon states [31], determining the degree of entanglement after devising appropriate Bell tests, and ultimately utilizing entangled x-ray probes to characterize entangled quantum materials.

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Data availability. There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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