

Emergence of Multiphoton Quantum Coherence by Light Propagation

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Cite This: *ACS Photonics* 2024, 11, 3197–3204



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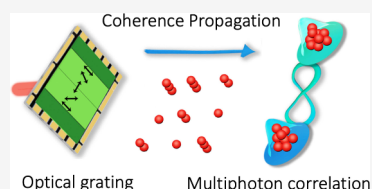
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ABSTRACT: The modification of the quantum properties of coherence of photons through their interaction with matter lies at the heart of the quantum theory of light. Indeed, the absorption and emission of photons by atoms can lead to different kinds of light with characteristic quantum statistical properties. As such, different types of light are typically associated with distinct sources. Here, we report on the observation of the modification of quantum coherence of multiphoton systems in free space. This surprising effect is produced by the scattering of thermal multiphoton wavepackets upon propagation. The modification of the excitation mode of a photonic system and its associated quantum fluctuations result in the formation of different light fields with distinct quantum coherence properties. Remarkably, we show that these processes of scattering can lead to multiphoton systems with subshot-noise quantum properties. Our observations are validated through the nonclassical formulation of the emblematic van Cittert–Zernike theorem. We believe that the possibility of producing quantum systems with modified properties of coherence, through linear propagation, can have dramatic implications for diverse quantum technologies.

KEYWORDS: Quantum coherence, Multiphoton, Nonclassical correlations, Quantum fluctuations, Quantum van Cittert–Zernike theorem



1. INTRODUCTION

The description of the evolution of spatial, temporal, and polarization properties of the light field gave birth to the classical theory of optical coherence.^{1–7} Naturally, these properties of light can be fully characterized through the classical electromagnetic theory.³ Furthermore, there has been interest in describing the evolution of propagating quantum optical fields endowed with these classical properties.^{8,9} This has been accomplished by virtue of the Wolf equation and the van Cittert–Zernike theorem.^{1,8,10,11} Nevertheless, there is a long-sought goal in describing the evolution of the inherent quantum mechanical properties of the light field that define its nature and kind.^{12,13} Such formalism would enable modeling the evolution of the excitation mode of propagating electromagnetic fields in the Fock number basis. Given the large number of scattering and interference processes that can take place in a quantum optical system with many photons, this ambitious description remains elusive.^{14–18} Although, it is essential to describe the evolution of propagating multiphoton wavepackets in diverse quantum photonic devices.^{18–22}

The quantum theory of optical coherence developed by Glauber and Sudarshan provides a description of the excitation mode of the electromagnetic field.^{12,13,23} This elegant formalism led to the identification of different kinds of light that are characterized by distinct quantum statistical fluctuations and noise levels.^{12,15,23–25} As such, a particular quantum state of light is typically associated with a specific emission process and a light source.²³ Moreover, the quantum theory of electromagnetic radiation enables describing light–

matter interactions.²⁶ These consist of absorption and emission processes that can lead to the modification of the excitation mode of the light field and consequently to different kinds of light.^{23,26} This possibility has triggered interest in achieving optical nonlinearities at the single-photon level to engineer and control quantum states of light.^{27–30} Thus, it is believed that the excitation mode of the light field, and its quantum properties of coherence, remain unchanged upon propagation in free space.^{23,26}

We demonstrate that the statistical fluctuations of thermal light fields and their quantum properties of coherence can be modified upon propagation in the absence of light–matter interactions.^{9,31} This effect results from the scattering of multiphoton wavepackets that propagate in free space. The large number of interference effects hosted by multiphoton systems with up to 20 particles leads to a modified light field with evolving quantum statistical properties.^{14,16} Further, we show that the evolution of multiphoton quantum coherence can be described by the nonclassical formulation of the van Cittert–Zernike theorem.⁹ Our description of propagating multiphoton quantum coherence unveils conditions under which multiphoton systems with subshot-noise quantum

Received: March 25, 2024

Revised: July 29, 2024

Accepted: July 30, 2024

Published: August 8, 2024



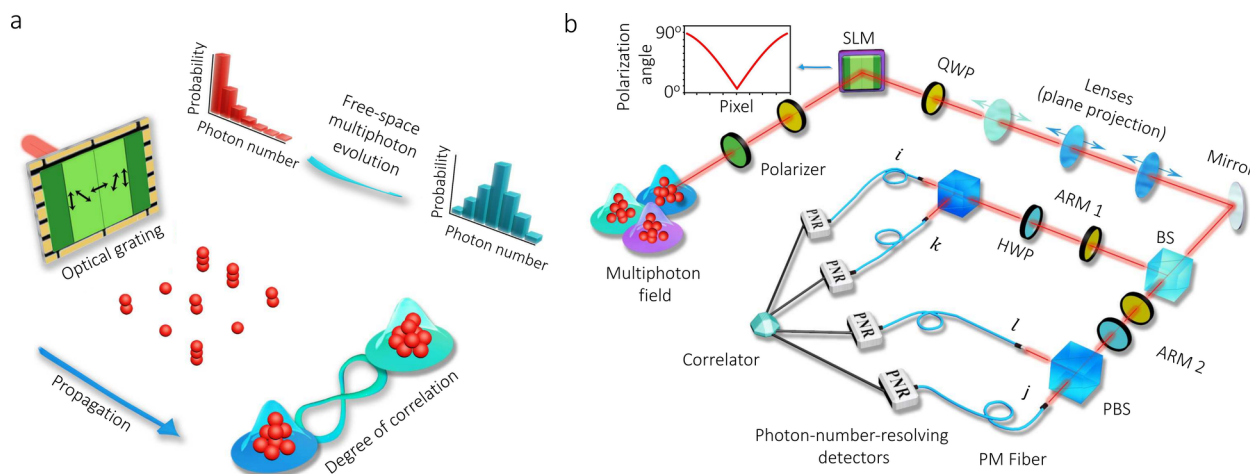


Figure 1. Modification of quantum coherence in propagating multiphoton wavepackets. The diagram in (a) illustrates the scattering of thermal multiphoton wavepackets by an optical grating. The grating modifies the polarization properties of the multiphoton wavepackets at different transverse spatial locations. The interference of the scattered multiphoton wavepackets, at different propagation planes, leads to changes in the quantum statistical properties of the thermal field. These interference events lead to the modification of multiphoton quantum coherence upon propagation in free space. The setup for the experimental investigation of this effect is shown in (b). Here, a multimode thermal multiphoton beam passes through a polarizer and a quarter-wave plate (QWP) to modulate its polarization. The transmitted circularly polarized photons illuminate a spatial light modulator (SLM) where we display a polarization grating. The beam reflected off the SLM is sent to another QWP to rotate its polarization at different transverse positions (details can be found in the SI).³⁵ The resulting polarization angle as a function of the transverse pixel position is depicted next to the SLM. This experimental arrangement induces partial polarization properties to the initial thermal light beam. The multiphoton field is then sent to a tunable telescope consisting of three lenses. This setup enables us to select different propagation planes of the scattered multiphoton field. We then perform polarization tomography of multiphoton wavepackets at an specific propagation plane by means of a beam splitter (BS), half-wave plates (HWPs), QWPs, and two polarizing beam splitters (PBS).³⁶ Finally, we use superconducting nanowire single-photon detectors (SNSPDs) to implement photon-number-resolving (PNR) detection as outlined in refs 24 and 37. This technique allows us to characterize the quantum coherence of propagating multiphoton systems and their quantum statistical fluctuations.

properties are formed.³² Remarkably, these quantum multiphoton systems are produced upon propagation in the absence of optical nonlinearities.^{27–30} As such, we believe that our findings provide an all-optical alternative for the preparation of multiphoton systems with nonclassical statistics. Given the relevance of photonic quantum control for multiple quantum technologies, similar functionalities have been explored in nonlinear optical systems, photonic lattices, plasmonic systems, and Bose–Einstein condensates.^{17,27–30,33,34}

2. EXPERIMENT AND RESULTS DISCUSSION

The optical system under consideration is depicted in Figure 1a. In this case, we utilize an unpolarized thermal field with a temporal coherence time of approximately 1 μ s, which is scattered by an optical grating to produce a multiphoton field with distinct polarization properties at different transverse spatial positions.⁵ Here, the thermal multiphoton field is produced by many independent wavepackets at the single-photon level with random relative phases.³⁸ The scattered photons contained in the thermal beam interfere upon propagation to change the statistical fluctuations of the field.⁹ These effects enable the modification of the quantum properties of coherence of the initial multiphoton thermal system in free space. As discussed in the Supporting Information (SI), we describe our initial thermal system as an incoherent superposition of coherent states^{12,13,39}

$$\hat{\rho} = \int d\Sigma \otimes_s (|\alpha\rangle\langle\alpha|_{\Sigma,H,s} + |\alpha\rangle\langle\alpha|_{\Sigma,V,s}) \quad (1)$$

where $|\alpha\rangle_{\Sigma,B,s}$ represents a coherent state of amplitude α with stochastically random mode-structure Σ , where $\hat{a}_{\Sigma,B,s} = \int d\rho \text{Rect}[(s - \rho)/d] \Sigma(\rho) \hat{a}_B(\rho)$ and polarization $B \in$

$\{H, V\}$ (see SI). Furthermore, the tensor product over positions s represents the pixelated transverse beam profile where each pixel has sidelength d .

After the polarization grating, the resulting state is obtained via the transformation

$$\hat{a}_B(\rho) \rightarrow P_{HB}(\rho) \hat{a}_H(\rho) + P_{VB}(\rho) \hat{a}_V(\rho) + P_{\emptyset B}(\rho) \hat{a}_{\emptyset}(\rho) \quad (2)$$

where $\hat{a}_{\emptyset}(\rho)$ is the mode for photon loss, and P_{AB} are the components of the transformation

$$P(x) = \begin{bmatrix} \cos^2\left(\frac{\pi x}{L}\right) & \cos\left(\frac{\pi x}{L}\right) \sin\left(\frac{\pi x}{L}\right) \\ \cos\left(\frac{\pi x}{L}\right) \sin\left(\frac{\pi x}{L}\right) & \sin^2\left(\frac{\pi x}{L}\right) \\ \sin\left(\frac{\pi x}{L}\right) & \cos\left(\frac{\pi x}{L}\right) \end{bmatrix} \quad (3)$$

where $A \in \{H, V, \emptyset\}$ and L is the length of the polarization grating. The beam described by eq 1 is then propagated by a distance of z before being measured by two pairs of photon-number-resolving (PNR) detectors.^{24,40} This propagation can be modeled through the Fresnel approximation on the mode structure of the initial beam.⁴¹ We can then compute the second-order correlation function $G_{ijkl}^{(2)}(r_1, r_2, z)$ for the post-selected polarization measurements in the detection plane as²

$$\begin{aligned} G_{ijkl}^{(2)}(r_1, r_2, z) &= \langle \hat{a}_i^\dagger(r_1) \hat{a}_j^\dagger(r_2) \hat{a}_k(r_1) \hat{a}_l(r_2) \rangle \\ &= I_0 \int d\rho_1 d\rho_2 d\rho_3 d\rho_4 F(r_1, r_2, \rho_1, \rho_2, \rho_3, \rho_4, z) \\ &\quad \times [\delta(\rho_1 - \rho_3) \delta(\rho_2 - \rho_4) + \delta(\rho_1 - \rho_4) \delta(\rho_2 - \rho_3)] \end{aligned} \quad (4)$$

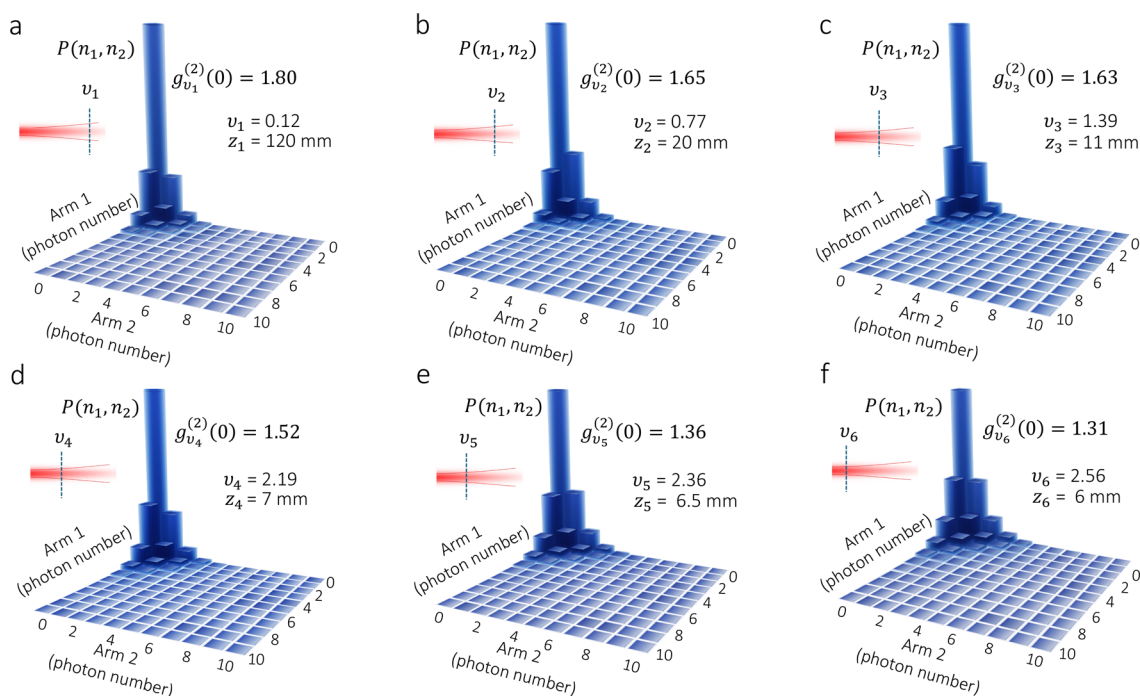


Figure 2. Evolving quantum coherence induced by light propagation. The propagation of the multiphoton system reflected off the SLM induces modifications in its photon-number distribution. In this case, we focus on the horizontally polarized component of the initial thermal beam with up to 20 particles. As shown in (a), the multiphoton system at the propagation plane characterized by $\nu_1 = 0.12$ is nearly thermal. We define ν as $L\Delta X/(\lambda z)$, in this case $L = 3$ mm, $\Delta X = 4$ mm, $\lambda = 780$ nm and we scan the propagation distance z . The average photon number we use in the detection plane, starting from the near field, is 1. The quantum fluctuations of the multiphoton system are attenuated with ν . This is quantified through the degree of second-order coherence $g_{\nu}^{(2)}(0)$, which also evolves upon propagation. The experimental results from a to f were obtained by scanning two detectors through different propagation planes. The large number of interference events upon propagation leads to the modified multiphoton system in f, which is characterized by a $g_{\nu_6}^{(2)}(0)$ of 1.31. This multiphoton beam exhibits quantum statistical properties that approach those observed in coherent light. The evolving quantum dynamics of our multiphoton system can be modeled through eq 5. Remarkably, the conversion processes of the multiphoton system and its modified properties of quantum coherence take place in the free space in the absence of light–matter interactions.

The term $F(r_1, r_2, \rho_1, \rho_2, \rho_3, \rho_4, z)$ accounts for the process of polarization preparation and field propagation (see SI). Moreover, I_0 is a normalization constant. Remarkably, the Dirac-delta functions in eq 4 give rise to the formation of nontrivial correlations in the propagated thermal light field. This nonseparability of the second-order coherence produces nonlocal correlations. The origin of these nontrivial correlations have been attributed to processes of two-photon interference.⁴² In our case, the delta functions $\delta(\rho_1 - \rho_3)\delta(\rho_2 - \rho_4)$ describe the coherence of a photon with itself. This kind of coherence existed prior to interacting with the grating. In addition, the delta functions $\delta(\rho_1 - \rho_4)\delta(\rho_2 - \rho_3)$ account for multiphoton spatial coherence gained by the system upon propagation. These terms unveil the possibility of modifying quantum coherence of multiphoton systems upon propagation.⁹ We use this approach to describe the correlation properties of the multiphoton wavepackets that constitute our light beam. This allows us to use an equivalent density matrix for the system $\hat{\rho}_{ijkl}(z)$ (see SI) at the detection plane to compute its corresponding joint photon-number distribution $p_{ijkl}(n_1, n_2, z)$ as

$$p(n_1, n_2, z) = \text{Tr}[\hat{\rho}_{ijkl}(z)|n_1, n_2\rangle\langle n_1, n_2|] \quad (5)$$

As we shall see in the next section, these formulas allow for the prediction of very interesting correlation effects. It is worth noticing that eq 5 describes projective measurements and not

conditional measurements as in schemes for photon-subtraction.^{43–46} Specifically, they predict that the statistical makeup of the light field is changing upon propagation in free space. Indeed, the classical analogue to this behavior is explained by the van Cittert–Zernike theorem,^{10,11} which predicts that the classical coherence properties of a light source can change upon free-space propagation. Therefore, we interpret our results in eq 4 as those of a quantum van Cittert–Zernike theorem. This is because they predict the modification of quantum coherence upon free-space propagation, and that is directly analogous to the classical theorem’s predictions. Specifically, eq 4 predicts this free-space quantum modification through the nontrivial scattering effects induced by the Dirac-delta functions. Furthermore, eq 5 allows us to study multiparticle quantum coherence, which is also changing upon free-space propagation. These quantum van Cittert–Zernike effects, therefore, are not only arising in polarization subsystems, but also in multiphoton subsystems. This showcases the fundamental and intrinsic quantum impacts of free-space propagation on our state.

We explore the modification of the quantum coherence properties of propagating multiphoton systems using the experimental setup in Figure 1b. We use a combination of waveplates and a spatial light modulator (SLM) to rotate the polarization properties of our multiphoton system at any transverse position.³⁵ In addition, this arrangement enables us to characterize the polarization and photon-number distribu-

tion of multiphoton systems at different propagation planes. Specifically, we perform measurements at different propagation planes associated with the propagation parameter $\nu = L\Delta X/(\lambda z)$. Here, the transverse distance between detectors is described by ΔX and the wavelength of the beam by λ . As demonstrated in Figure 2, the many interference effects hosted by the propagating multiphoton system modify the photon-number distribution of the polarized components of the initial beam.^{15,17,20,47} These processes lead to multiphoton systems with different quantum fluctuations and quantum properties of coherence.^{15,48} Each multiphoton system is characterized through the degree of second-order self-coherence

$$g_{\nu}^{(2)}(0) = \frac{G_{\text{HHHH}}^{(2)}(\mathbf{r}, \mathbf{r}, z)}{G_{\text{HH}}^{(1)}(\mathbf{r}, z)^2} \quad (6)$$

where $G_{i,j}^{(1)}(\mathbf{r}, z) = \langle \hat{a}_i^\dagger(\mathbf{r}) \hat{a}_j(\mathbf{r}) \rangle = \sqrt{I_0} L / (2z^2 \lambda^2)$. The multiphoton system in Figure 2a is nearly thermal.²³ However, propagation leads to different kinds of multiphoton wavepackets. We show these from Figure 2a to f. The coherence properties of the multiphoton system in Figure 2f approach those observed in coherent light beams.³² While the magnitudes of the degree of coherence in Figure 2 are typically attributed to classical properties of the light field, these properties result from nonclassical interference processes as described by eq 4. As such, these same processes of coherence can also produce nonclassical statistics that lead to degrees of second-order coherence below one. We discuss these effects below. Remarkably, the modification of the multiphoton system in Figure 2 takes place in free space, in the absence of light–matter interactions.^{17,26–29,33,34}

The polarization and photon-number properties of the propagating light beam at different transverse and longitudinal positions host many forms of interference effects.^{14,39} We explore these dynamics by isolating the constituent multiphoton subsystems of the propagating beam. Each multiphoton subsystem, characterized by different polarization properties, exhibits different degrees of second-order coherence.²⁰ In the experiment, we perform projective measurements on polarization. These measurements unveil the possibility of extracting multiphoton subsystems with attenuated quantum fluctuations below the shot-noise limit.^{37,40} In this case, we use the four detectors depicted in the experimental setup in Figure 1b to perform full characterization of polarization.³⁶ These measurements enable us to characterize correlations of multiphoton subsystems with different polarization properties, which are reported in Figure 3. We plot the degree of second-order mutual coherence

$$g_{ijkl}^{(2)}(\mathbf{r}_1, \mathbf{r}_2, z) = \frac{G_{ijkl}^{(2)}(\mathbf{r}_1, \mathbf{r}_2, z)}{G_{i,j}^{(1)}(\mathbf{r}_1, z) G_{k,l}^{(1)}(\mathbf{r}_2, z)} \quad (7)$$

The propagation of the multiphoton subsystem described by $g_{\text{HHHH}}^{(2)}$ shows a modification of the quantum statistics from super-Poissonian to nearly Poissonian.^{32,48} A similar situation prevails for the multiphoton subsystem described by $g_{\text{HHHV}}^{(2)}$. It is worth noticing that the multiphoton subsystems described by $g_{\text{VHHV}}^{(2)}$ and $g_{\text{HHVV}}^{(2)}$ unveil the possibility of extracting multiphoton subsystems with subshot-noise properties.⁴⁹ Notably, the $g_{\text{HHHV}}^{(2)}$ measurement involves the annihilation and creation of H photons at one detector, and the annihilation and creation of V photons at a second detector.

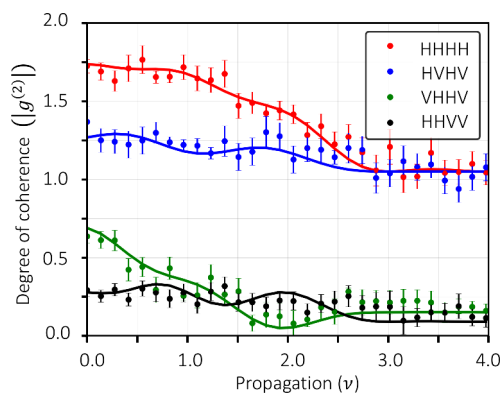


Figure 3. Measurement of multiphoton light with subshot-noise properties. We isolate multiphoton subsystems with different polarization properties. These are characterized by the degree of second-order coherence $g_{ijkl}^{(2)}$. While the four multiphoton subsystems indicate the modification of quantum coherence with the ν parameter, it should be highlighted that the subsystems described by $g_{\text{VHHV}}^{(2)}$ and $g_{\text{HHVV}}^{(2)}$ show degrees of coherence below one. Notably, quantum light sources with quantum statistical fluctuations below the shot-noise limit show degrees of coherence smaller than one. The continuous lines represent our theoretical predictions from eq 4, whereas the dots indicate experimental data.

This measurement can be implemented classically, resulting in $g_{\text{HHHV}}^{(2)} > 1$. Conversely, the $g_{\text{VHHV}}^{(2)}$ measurement involves the annihilation and creation of a photon with different polarizations at each detector. This process cannot be described using a unitary operator, therefore, it does not constitute a classical measurement. The nonclassical behavior of this multiphoton subsystem is quantified by $g_{\text{VHHV}}^{(2)} < 1$, which confirms the presence of quantum coherence. This implies photon-number distributions narrower than the characteristic Poissonian distribution of coherent light.^{23,50} The red trace in Figure 3 indicates an evolution of the multiphoton subsystem from super-Poissonian to Poissonian as depicted in Figure 1a. This peculiar feature might unlock novel paths toward the implementation of sensitive measurements with subshot-noise fluctuations.⁵¹

We now turn our attention to describe the quantum coherence evolution of propagating multiphoton wavepackets. This is explored by projecting the polarized components of the initial thermal beam into its constituent multiphoton wavepackets.²³ In this case, we analyze wavepackets with $n_1 + n_2$ number of photons. The number of photons detected in arm 1 of our experiment is described by n_1 , whereas n_2 indicates the number of photons detected in arm 2. Our findings unveil that despite the fact that the degree of second-order coherence $g_{\text{HHHH}}^{(2)}$ in Figure 3 is produced by its constituent wavepackets, these show a completely different evolution of their properties of coherence. Our experimental measurements of these wavepackets can be found from Figure 4a–c. The results in Figure 4a indicate that multiphoton wavepackets, in which n_1 and n_2 are the same, show a particular evolution. In contrast, propagating wavepackets with asymmetric values of n_1 and n_2 show different trends in the modification of quantum coherence, these are shown in Figure 4b,c. The propagation of these wavepackets can be described using eq 5. Specifically, we can calculate the multiphoton degree of second-order mutual coherence⁵²

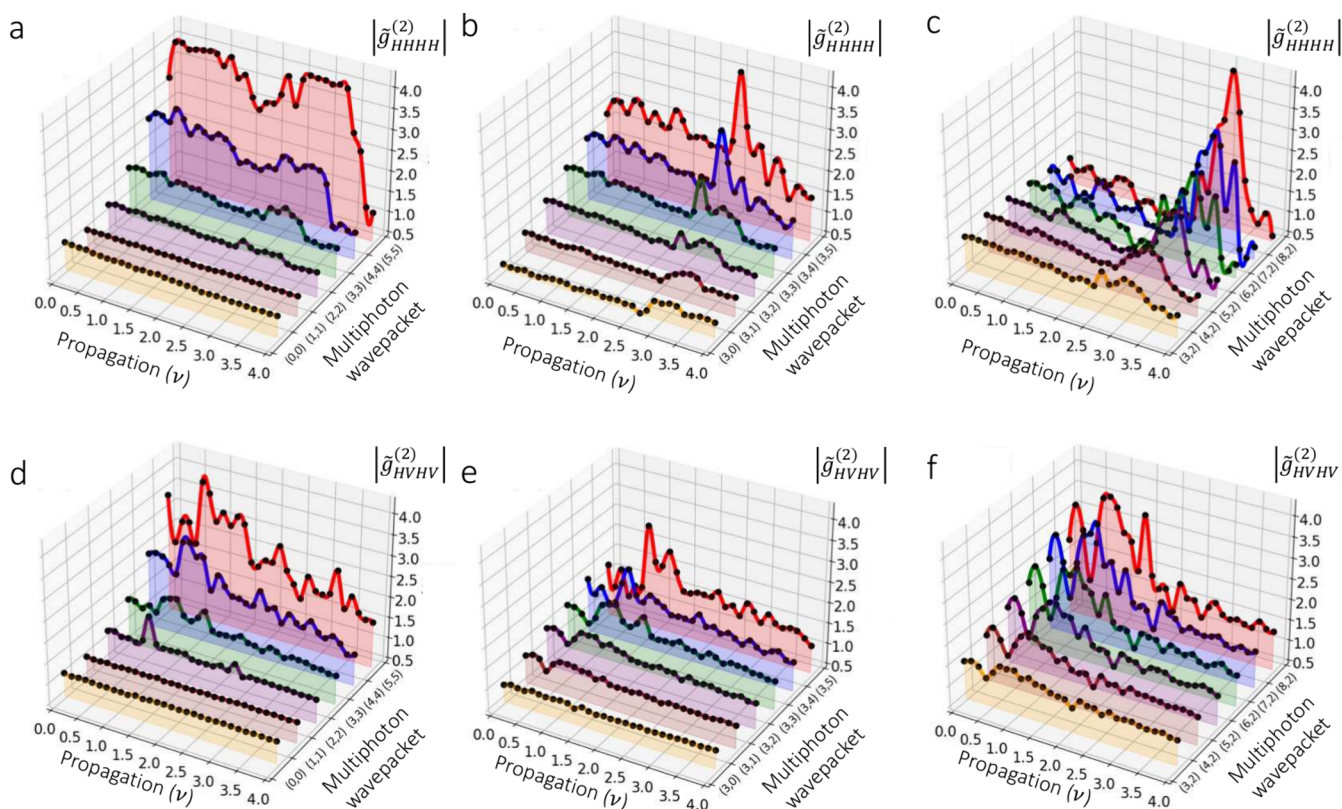


Figure 4. Quantum coherence of propagating multiphoton wavepackets. The panels from (a) to (c) show the evolution of multiphoton wavepackets contained in the horizontally polarized component of the initial thermal beam. We label the multiphoton wavepacket that leads to the detection of n_1 photons in arm 1 and n_2 photons in arm 2 with (n_1, n_2) . The results from (a) to (c) indicate that the multiphoton events that produce the degree of second-order coherence $|\tilde{g}_{HHHH}^{(2)}|$ in Figure 3 follow distinct propagation dynamics. Although the contributions from the constituent wavepackets produce the trace described by $|\tilde{g}_{HHHH}^{(2)}|$, their individual propagation shows different coherence evolution. Specifically, we identify three representative dynamics. For example, multiphoton wavepackets with equal numbers for n_1 and n_2 exhibit the propagation dynamics in (a). In contrast, propagating wavepackets with different values of n_1 and n_2 show a different trend for the modification of quantum coherence, these are shown in (b) and (c). Moreover, the multiphoton wavepackets in the projected beam characterized by $|\tilde{g}_{HVVH}^{(2)}|$ exhibit the multiphoton dynamics reported from (d) to (f). The multiphoton dynamics in these panels also depend on the number of photons in each of the measured wavepackets.

$$\tilde{g}_{ijkl}^{(2)}(n_1, n_2, z) = \frac{p_{ijkl}(n_1, n_2, z)}{\sum_n p_{ijkl}(n, n_2, z) \sum_m p_{ijkl}(n_1, m, z)} \quad (8)$$

Furthermore, the multiphoton wavepackets in the projected beam, characterized by $\tilde{g}_{HVVH}^{(2)}$, exhibit the multiphoton dynamics reported from Figure 4d–f. These results suggest that the multiphoton dynamics in Figure 4 depend on the number of photons in each of the measured wavepackets.

This quantum field theoretic approach to studying the quantum van Cittert–Zernike theorem provides us with the ability to describe the propagation dynamics of the multiphoton systems that constitute classical light beams. We used this formalism to extract propagating multiphoton subsystems, with quantum statistical properties, from unpolarized thermal light fields. While nonlinear light–matter interactions offer the possibility of engineering complex quantum systems,^{27–29} our scheme exploits linear propagation of multiphoton systems^{30,53} and the nonlinearity of projective measurements on photon-number states.^{30,53} This feature enabled us to exploit multiphoton scattering in free space to produce wavepackets with different quantum statistical properties.¹⁵ As such, our work combines the benefits of postselective measurements

with those of multiphoton scattering in propagating light beams, and it allows us to study the modification of the quantum statistical properties of multiphoton wavepackets in free space. Although, the incoherent combination of light beams with different polarization properties can lead to the modification of the degree of second-order coherence,^{17,31} we performed direct measurements of polarized multiphoton systems with propagating quantum coherence properties (see Figure 4a to c). These processes are defined by the number of particles in the measured multiphoton system. Consequently, these findings have important implications for all-optical engineering of multiphoton quantum systems.

The formulation of the van Cittert–Zernike theorem has played a fundamental role in the development of optical instrumentation for astronomy.² Recently, there has been an enormous interest in the development of quantum-enhanced technology for astronomy.^{54–56} In this context, we believe that the emergence of multiphoton quantum coherence through light propagation, as described by the quantum van Cittert–Zernike theorem, holds significant implications for quantum astrometry and the advancement of quantum telescopes.^{54,55} Building on this foundation, the underlying physics behind the quantum van Cittert–Zernike theorem has been exploited to

develop robust quantum imaging technologies relying on the use of sun light.⁵⁷ Additionally, it is important to highlight the potential of using the dynamics reported in Figure 4 to demonstrate synthetic lattices in the photon-number basis.⁵⁸ This is relevant given the challenges associated with the demonstration of photonic lattices in the particle basis.^{59,60}

3. CONCLUSION

We demonstrated the possibility of modifying the excitation mode of thermal multiphoton fields through free space propagation. This modification stems from the scattering of multiphoton wavepackets in the absence of light–matter interactions.^{17,26–29,33,34,61} The modification of the excitation mode of a photonic system and its associated quantum fluctuations result in the formation of different light fields with distinct quantum coherence properties.^{12,13,23} The evolution of multiphoton quantum coherence is described through the nonclassical formulation of the van Cittert–Zernike theorem, unveiling conditions for the formation of multiphoton systems with attenuated quantum fluctuations below the subshot-noise limit.^{40,51,62} Notably, these quantum multiphoton systems emerge in the absence of optical nonlinearities, suggesting an all-optical approach for extracting multiphoton wavepackets with nonclassical statistics. We believe that the identification of this surprising multiphoton dynamics has important implications for multiphoton protocols quantum information.^{18,21}

■ ASSOCIATED CONTENT

Data Availability Statement

The data sets generated and/or analyzed during this study are available from the corresponding author or last author on reasonable request.

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsp Photonics.4c00558>.

Description of the unpolarized multimode thermal light, the propagation of the light to the farfield, the second order correlation of the propagated beam, the photon number distribution of the propagated beam, and the realization of polarization grating (PDF)

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Funding

M.H., R.B.D., F.M., C.Y., and O.S.M.L. acknowledge support from the Army Research Office (ARO), through the Early Career Program (ECP) under the Grant No. W911NF-22-1-0088. J.F. acknowledges funding from the National Science Foundation through Grant No. OMA 2231387. A.O. acknowledges funding from the National Science Foundation through Grant No. DMS 2206037. R.J.L.-M. thankfully acknowledges financial support by DGAPA-UNAM under Project UNAM-PAPIIT IN101623.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank Dr. James A. Joseph for very useful discussions on the quantum control of multiphoton dynamics.

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