

Applications of single photons in quantum metrology, biology and the foundations of quantum physics

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Abstract

With the development of photonic quantum technologies, single photons have become key for various applications including quantum communication and quantum computing, discussed in an accompanying Review. Here we overview the applications of single photons in quantum metrology, biology and experiments probing the foundations of quantum physics. For each of these applications, we outline the main milestones reached so far, the remaining challenges, and the improvements that could be made in the future. We conclude with a wish list for future single-photon sources.

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Outlook

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Key points

- Detecting a single-photon or an N-photon state is not easy and requires specific detectors with precise calibration.
- Quantum metrology with single photons can reach the ultimate limit in terms of metrology standards.
- Natural biological systems, such as the human eye and processes in photosynthesis, are sensitive to the detection of single photons.
- Single-photon experiments are used to test the limits of quantum mechanics and uncover connections to other theories such as general relativity.

Introduction

We continue the discussion of applications of single photons that we started in the accompanying Review¹ and also direct the readers to other existing reviews^{2–6}. This Review is complemented by further information provided in the Supplementary Information and, where necessary, we direct the reader to it.

Applications in quantum metrology

Introduction to quantum metrology

The detection of single photons is an enabling technology for many applications because single-photon detection with low dark counts allows measurements at the shot-noise limit (see the Supplementary Information); thus, the photon-number detection uncertainty is not limited by background noise, but by the photon statistics of the evaluated state itself. Measuring the photon rate in a light-starved environment or measuring the time of flight are mature methodologies using single photons for improving measurement accuracy or decreasing the overall measurement time. However, losses limit most of these applications, preventing an advantage from using quantum effects that can result in quantum-enhanced performance; this is because loss results in a noisier measurement, therefore eliminating the quantum advantage. Hence, the most important task is to limit the total system loss. We define loss as $(1 - \eta)$, where η is the overall probability of detecting a photon after its creation at the source; that is, the overall transmission or ‘efficiency’. One can mitigate the noise due to loss $(1 - \eta)$ by using a coincidence-based post-selection protocol (see the Supplementary Information). This method is based on the heralded single-photon source obtained from photon-pair production coming from spontaneous parametric down-conversion (SPDC), described in ref. 1 and its supplementary information. The trade-off is an increase in the overall measurement time (t_m), as a post-selected measurement requires the desired state (that is, number of detected photons N) to be present at the detection unit. This time scales exponentially with the number of photons in the desired state, $t_m \propto \eta^N$. In this case, the total system loss is relevant and cannot be ignored by post-selection as undetected photons may still have interacted with the sample. In addition, noise contributions due to background photons or dark counts at the detection unit will increase a minimum loss requirement to achieve quantum advantage, as these spurious detection events increase the overall noise detected by the system.

Figure 1a shows an ideal single-photon source (single-photon gun), in which precisely one photon at a time is emitted. In contrast, a weak coherent laser emits a random number of photons at a given time

interval. Figure 1b shows the standard deviation (in photon number) of the counting statistics as would be measured by a photon-number resolving detector (σ_N in photon number; see the Supplementary Information) as a function of overall system loss (η) for both photon sources. This standard deviation represents the shot-to-shot uncertainty in detected photon number for the given state of light. Clearly, the single-photon gun ($\sigma_N = 0$ at $\eta = 1$) has a huge advantage over a weak coherent laser with mean photon number 1.

Needs and requirements

In quantum metrology with photons, four aspects are studied and investigated for applications: high-efficiency detection, photon-number resolution, quantum-enhanced resolution and time-of-flight measurements. These will be described below.

High-efficiency detection. The total system efficiency of a single-photon source (SPS) is desired to be close to unity¹. If combined with on-demand (push-button) operation, selectable wavelength of operation and indistinguishability of the generated photons, such a source–detector system would be the ultimate tool for single-photon metrology, and excellent progress towards on-demand operation has been made^{7–10}. Ultimately, an SPS is desired in which one can pick and choose the number of photons in a certain pulse of light with unity detection efficiency. For example, such an N -photon Fock state will obey photon statistics ruled by a binomial distribution, rather than a Poisson distribution (such as that of a weak coherent laser), as illustrated in Fig. 1a for a Fock state of $N = 1$. As a result, this N -photon Fock state will display a smaller variance in the photon statistics than a Poisson distribution when experiencing the same loss. Therefore, fewer experimental trials (or shorter measurement times) are required to deduce, for instance, the transmission of an object using such an N -photon Fock state as compared with a coherent state of the same mean photon number $\langle N \rangle$ (refs. 11–13).

Photon-number resolution. For many optical quantum information applications based on post-selective coincidence detection (see ref. 1), it is enough to be able to detect the resulting output state with detectors capable of discriminating zero from one, two or more photons. A click detector (that is, a single event recorded on a photon detector) serves part of this requirement. However, being able to discriminate two or more photons from one photon is in principle not possible using a single click detector. Below, we will describe ways to discriminate photon numbers using click detectors.

Quantum-enhanced resolution. Improved sensitivity through measurements using quantum systems is one of the first practical applications of quantum technologies such as better sensors and measurement systems to improve resolution. To achieve this, research focuses on how to use optical quantum states to improve length, spatial, temporal and spectral resolution. A prime example is the quantum-enhanced measurement of phase (length) using squeezed vacuum^{14–16}. Here, the noise of one electric field quadrature is squeezed in a way that allows a homodyne measurement to perform a better phase measurement (or equally accurate phase measurement using fewer photons) than using a coherent laser beam operating at the standard quantum limit. Another example is an enhanced optical microscope using entanglement, as developed in ref. 17.

Time-of-flight measurements. When a time-stamped photon source is used (such as a pulsed laser source), the timing between the photon

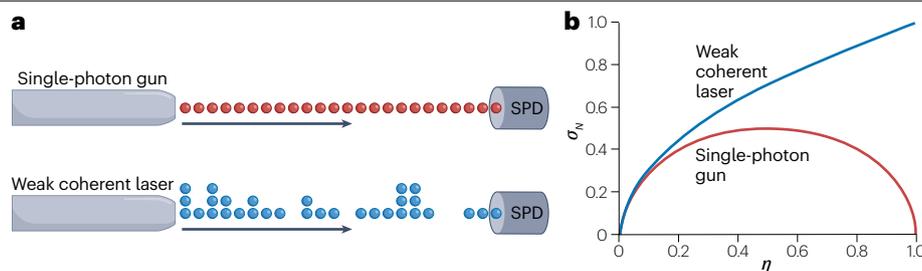


Fig. 1 | Difference between a coherent and a Fock state. a, Photons emitted from an ideal single-photon source (single-photon gun) – a regular photon emission with one photon at a time (top). Photons emitted from a weak coherent laser according to a Poisson distribution – the photon emission and the number of

photons are random (bottom). **b**, The standard deviation (in photon number) of the counting statistics measured by a photon-number resolving detector (σ_n) as a function of overall system loss (η). SPD, single-photon detector.

generation and detection can give insight about the photon's travel time. In some applications, this 'time-of-flight' measurement allows for depth estimation, that is, the travel time of a photon reflecting off a target. For example, 'self-driving' cars require scanning their immediate environment to make appropriate decisions in almost real time. The 3D scanning is done with a time-of-flight LiDAR system using short laser pulses and avalanche photodiodes¹⁸. To achieve best spatial resolution, a low-timing-jitter detector is desired, a detector with low uncertainty in timing between the photon absorption event and detected electrical signal.

Achieved milestones

Numerous developments have been reported in the context of quantum metrology. In the following, we will again focus on four aspects: high-efficiency detection, photon-number resolution, quantum-enhanced resolution and time-of-flight measurements.

High-efficiency detection. In the past decade, tremendous improvements in single-photon detector performance have been made. Efficiencies can now be more than 95% (refs. 19–21), and dark counts can be as low as a few counts per day²². However, these detectors require a cryogenic environment to operate, and the photons are generally fibre-coupled to the active area of the detector. To achieve low system losses, the task is therefore to couple a quantum state of light efficiently into an optical fibre. Many groups have succeeded in achieving high-efficiency coupling of an SPS into fibre^{10,23–26}. So far, a record efficiency of 57% from an SPS to a single-mode fibre has been achieved, without including the detector efficiency and coupling. Once the efficiency of the detector and the coupling to it is added, this value will go down, showing the importance of ongoing optimization of the overall efficiency of an SPS from creation to detection. Ideally, the product of the detector efficiency and source efficiency should be greater than 2/3, which seems close to what is currently possible²⁷.

Photon-number resolution. Recent progress suggests that by evaluating more detector response parameters beyond a simple threshold ('click'), some photon-number resolution can be recovered when using superconducting nanowire single-photon detectors²⁸ (see also ref. 1). An alternative method to achieve photon-number resolution is by use of click detector multiplexing in the spatial^{29,30} or temporal domain³¹. These approaches result in a quasi-photon-number resolving capability of the detector. The detector outcome does not perfectly resemble the optical state that is measured, but it reduces the probability of

error due to multiphoton detection events in one of the detector elements while increasing the number of detector elements³². A truly photon-number-resolving detector is the optical transition-edge sensor (TES), which has been used in a variety of quantum optics and quantum metrology experiments³³. Operating in the transition between the superconducting and normal regime, the TES's active area resistance changes owing to heating after absorption of a pulse of energy: that is, the resistance change depends on the number of photons absorbed by the active area. In the visible spectral range, the typical maximum number of photons discernible from the TES is around 15, after which saturation effects wash out the capability to resolve single-photon numbers^{34,35}. However, even in the saturated regime, a photon number can still be assigned, albeit with larger uncertainties³⁶. The TES can be particularly useful when accurately measuring the second-order correlation function $g^{(2)}$ (see ref. 1) or higher-order correlation functions, allowing the extraction of optical modes within the measured pulse of light^{37,38}. As an example, $g^{(2)}$ is exactly described by³⁹ $g^{(2)} = \sum_{n=0}^{\infty} N(N-1)p_n / (\sum_{N=0}^{\infty} Np_N)^2$, where N is the photon number and p_N is the photon-number probability of the measured state. Both of these parameters can be accurately measured with the TES. In contrast, when using click detectors in a Hanbury Brown and Twiss configuration⁴⁰, as described in Fig. 1 in ref. 1, the $g^{(2)}$ can only be approximated by $g^{(2)} \approx 2p_2/p_1^2$, where $p_1 \gg p_2 \gg p_3 \dots$ and p_1 is the probability of measuring one photon (approximately the mean photon number), whereas p_2 is the probability of measuring two photons coincidentally. Thus, this approximation using click detectors only holds for a low overall system efficiency or a system with very low higher-order photon probability. Being able to generate an N -photon state and to detect N single photons would be very useful for quantum metrology, but also for quantum computation as photon-number-resolved detection introduces an effective nonlinearity^{41,42}.

Quantum-enhanced resolution. One of the most prominent applications of squeezed light for enhanced phase resolution is the detection of gravitational waves in the Laser Interferometer Gravitational-Wave Observatory (LIGO). Squeezed light is injected into the interferometer, allowing for lower noise and higher displacement measurement accuracies. In addition to the squeezed vacuum state of light, the exploration of new and exotic quantum states of light is an important aspect of quantum metrology and optical quantum information processing.

Examples of such exotic states include coherent state superpositions – also called optical Schrödinger cat states^{43–46}. Coherent state superpositions are challenging optical quantum

states to generate as they require a quantum memory and iterative schemes. So far, only approximations have been realized through photon subtraction from a squeezed vacuum. Although these states can be used in linear optical quantum computing⁴⁷ and enhanced phase estimation protocols⁴⁸, their generation rate is probabilistic in nature because of the photon subtraction process, and therefore typically long measurement times are required. The same is true for phase measurements using NOON states⁴⁹ and the slightly more loss-tolerant variant of Holland–Burnett states⁵⁰ based on SPDC. In addition, some experimental demonstrations have made use of post-selection, which usually requires more resources, but this is not desirable because the samples used can be damaged through over-exposure. Accounting for all photons passing through the sample and comparing their number to the best classical estimator (weak coherent state) is the fairest evaluation of the photon state's enhanced performance. An unconditional measurement of quantum-enhanced phase estimation was demonstrated, using SPDC while obtaining above-classical Fisher information per detected photon⁵¹. This specific study used click detectors and was not able to distinguish between one or more photons. The use of photon-number-resolving TESs allows a larger quantum advantage over a much larger range of phase values⁵². However, the downside of these experiments is that the protocol implementations were based on SPDC, where the generation rate of the desired state is low and probabilistic. This may have detrimental consequences when fast phase estimation at the quantum limit is a requirement. Other emerging technologies are based on quantum-enhanced imaging protocols, where the phase, in addition to intensity of single-photon emitters, is measured either to beat Rayleigh's curse⁵³ or to beat Taylor's criterion⁵⁴. These newly developed protocols and demonstrations may find use in bio-imaging and chemistry.

Time-of-flight measurements. The most prominent application for time-of-flight/LiDAR measurement is the self-driving car. A pulsed, rotating (eye-safe) laser illuminates the scene around the car, and the reflected signal is picked up by a single-photon detector operating, ideally, at room temperature. This application drives the research and development to produce cheap (arrays of) single-photon detectors with low jitter and high dynamic range. Therefore, tremendous improvements in packaging and functionality have been achieved. On the other end of the spectrum, an extremely low-jitter detector has been demonstrated based on superconducting technology⁵⁵, unfortunately not yet near room temperature. Although this technology is still far from being mass-produced, it points a way to extremely high-precision timing measurements of single photons of the order of picoseconds, allowing for more exciting discoveries to be made in the future.

Future improvements

As already discussed in ref. 1, an emerging field in quantum technologies is the development of optical quantum networks. These networks will need components based on photonic quantum technologies. No matter the design of the future quantum nodes and components, photons will be used to transmit the quantum information on global scales. Therefore, robust, accurate and convenient tools for characterization of the transmitted input states and characterization of the component responding to an input state are prerequisites. The vast number of existing and emerging applications of single-photon counting will require the metrology tools for validation and benchmarking of systems and system components in the future. In the Supplementary Information, we describe single-photon detector calibration

methods that can serve as a baseline characterization tool for such optical quantum network components.

Applications in biology

Introduction to using single photons in biology

In recent years, single-photon sources have also become enabling tools for biological research, with a high potential for future applications. Light harvesting systems, such as the photoreceptor cells in the visual system or photosynthetic pathways, represent intriguing biological systems to study light–biomatter interaction at the single-quantum level. For this, the controlled generation of photonic states of different properties, both in space and time, is an essential prerequisite.

Needs and requirements

Most experimental approaches used in the past to study the interaction of light with biological systems relied on attenuated classical light sources, which obey Poissonian statistics. This, however, comes with considerable and irreducible statistical variability in the actual number of photons emitted and thus fundamentally prevents studies on the absolute (bio-)physical limits of light–matter interactions.

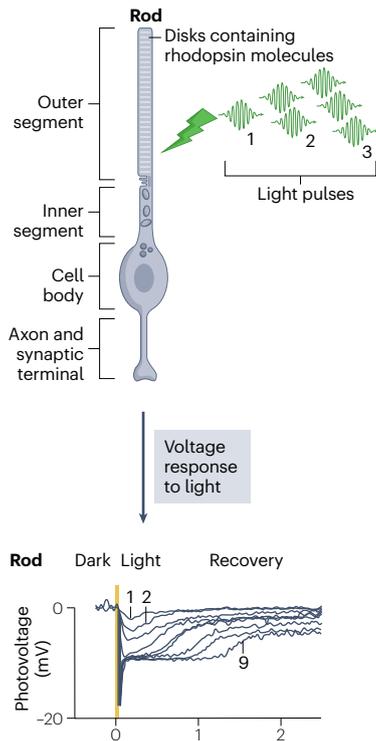
Light interacts with biological systems in many ways and is essential for life as we know it. Although most light–matter interaction takes place at macroscopic amplitudes, thereby involving the effect of an incoherent summation of thousands to billions of photons, there are a few particular cases in which the photon statistics of light do matter in biology.

One of the most striking examples is the single-photon response of retinal rod cells^{56,57}. The retina is a light-sensitive tissue that converts incident light into electrical signals, which are then transmitted along the visual pathway to higher processing centres of the brain⁵⁸. Experiments over the past 70 years^{59–61} have suggested that rod photoreceptors can detect light at the level of a few single photons with remarkable reproducibility and robustness to noise⁶², and thus represent miniaturized photodetectors which can, in principle, outperform several artificial photodetection devices. The photosensitive elements of rod cells are rhodopsin molecules which convert impinging light through an elaborate photochemical transduction pathway⁵⁶ (isomerization cascade) that eventually results in the hyper-polarization of the cell membrane, which constitutes an electrical signal (~ 60 mV) that can be further processed by the downstream cellular network of the vertebrate retina (Fig. 2a). A major question in the field pertains to understanding the biophysical processes that ensure the reliable detection and amplification of a single rhodopsin molecule response in the presence of considerable intrinsic biological (chemical and neural) noise at all stages of the visual system⁶³. Here, the application of well-controlled (single-photon) light sources provides a unique opportunity, as they aid in removing the 'input' noise that is generated by classical Poissonian light sources.

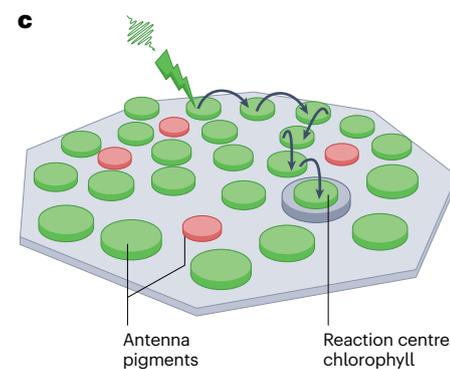
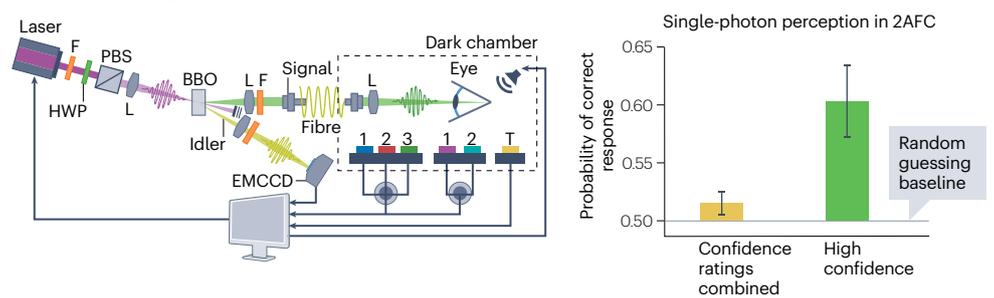
Achieved milestones

The advent of SPSs that possess tunable photon statistics has provided new ways to investigate the fundamental limit of photon detection and whether humans can indeed detect light intensity on the level of single quanta ($\sim 4 \times 10^{-19}$ J). Exploiting heralded single-photon emission through SPDC, the study in ref. 64 proved the single-photon sensitivity of frog rod photoreceptor cells and measured their quantum efficiency ($\sim 33\%$). In related work, the same group also investigated the impact of photon fluctuations of various classical light sources on their electrophysiological response by using coherent and pseudothermal light sources⁶⁵.

a Morphology of photoreceptors



b Detection of single photons by humans



d Correlated two-photon microscopy

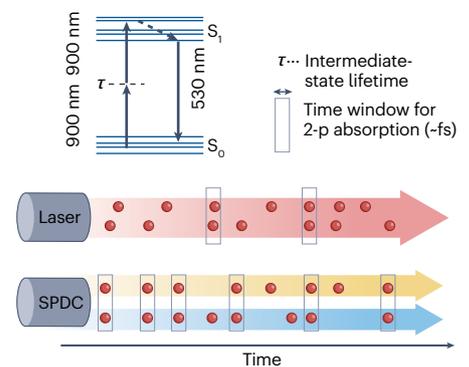


Fig. 2 | Application of single-photon sources in biology. **a**, The morphology of a rod cell and its response to light. Rhodopsin molecules in the outer-segment disks absorb single or multiple photons, which eventually leads to voltage responses of the cell which increase with the intensity of the light stimulation. **b**, The left side shows the schematics of the experimental set-up used to probe the absolute limit of light perception in humans. A non-degenerate spontaneous parametric down-conversion (SPDC) source supplies single photons to the experimenter. Here an electron-multiplying charge-coupled device (EMCCD) camera ensured rejection of multiphoton emission events via post-selection and that only single photons were further considered for analysis. BBO, beta barium borate; F, filter; HWP, half-wave plate; L, lens; T, transmitter. The graph on the right side shows the probability of providing the correct response in experimental trials for all post-selected single-photon events, demonstrating the above-chance probability for humans to consciously perceive single photons.

2AFC, two-alternative forced choice. **c**, Schematic of energy transfer via light-harvesting complexes involved in photosynthesis. The energy transfer between individual antenna pigment complexes involves quantum coherence whose properties could be further probed with single-photon input to the pathway. **d**, Correlated photon sources could enable efficient two-photon (2-p) excitation of fluorescence in microscopy. Two photons of longer wavelength excite a two-photon transition in the molecule if they arrive within the intermediate-state lifetime τ . The lower part shows the photon statistics of a laser versus SPDC photons. Photons produced in SPDC are highly correlated in time and could enhance the efficiency of the two-photon excitation by many orders of magnitude, consequently reducing photodamage and toxicity of the biological sample during imaging. Panel **a** adapted with permission from ref. 118, McGraw-Hill; panel **b** adapted from ref. 66 under a Creative Commons licence CC BY 4.0; panel **c** adapted from ref. 119, American Chemical Society.

The question of whether humans can unambiguously detect single photons was addressed in another study⁶⁶ (Fig. 2b). The experiment⁶⁶ also exploited post-selected SPDC photons that were produced in a degenerate fashion such that the signal photon's wavelengths overlapped with the peak of the spectral response of human rod cells (~500 nm) while the idler (~560 nm) coincided with the maximum quantum efficiency of their detector. One drawback of any heralded single-photon source based on SPDC is the inherent background due to multiphoton emission. Therefore, the authors⁶⁶ used an electron-multiplying CCD (EMCCD) camera for detecting the idler photon, which allowed them to identify and reject events in which more than one photon pair was generated, with higher efficiency than single-pixel single-photon avalanche diodes. This, in combination with an improved psycho-physics protocol (by using the 'two-alternative forced choice' method where the person had to say 'yes' or 'no' on the detection of a single photon, as well as give a confidence rating), allowed them to show

that subjects could detect a single photon with a probability above chance (Fig. 2b). Moreover, the study found a photon-induced temporal modulation of the visual system's gain, such that at the single-photon level, the detection probability of obtaining a photon was higher if another photon had been previously absorbed on the timescale of a few seconds.

The studies discussed above, enabled by a new generation of advanced single-photon sources, call for further studies on the fundamental biophysical limits of vision; they might also enable new experiments in quantum foundations and quantum biology⁶⁷. From a biological perspective, SPSs will enable a wide range of future studies. The precise spatial and temporal control of these sources could be used to study integration mechanisms in the rod visual system, for instance to measure temporal summation (the potential response to external signals for specific cells, such as neurons) in human subjects in the scotopic (low-light) condition^{68,69}. More sophisticated experiments in

animal models where the various cell layers in the retina are accessible for recording in ex-vivo preparations could allow the investigation of the biological mechanisms with which the complex retina neural circuitry reliably transmits and amplifies⁶³ the photoinduced electrical signals in the rod pathway, and how the integration of signals stemming from different spatial (and temporal) regions is performed (for sources that can generate more than one photon on demand). From a biophysical perspective, the fact that an SPS can reduce the ‘input’ noise into the visual system could enable experiments to better elucidate the cellular mechanisms that ensure the visual system’s remarkably high efficiency, accuracy and reproducibility, and to better characterize the various intrinsic noise sources⁷⁰. This may help to answer how biological signal detection at the absolute physical limits occurs in general^{71–75}.

The visual system is not the only biological system that operates on the single-photon level. The photosynthetic pathway⁷⁶ responsible for converting light into chemical energy, such as photosynthesis in plants, provides another area of ongoing biophysical research whose studies could tremendously benefit from the advances of reliable and well-controlled generation of single-photon states. The initial steps of photosynthesis comprise the absorption of sunlight by pigment-protein antenna complexes followed by rapid and highly efficient funnelling of excitation energy to a reaction centre (Fig. 2c). In these transport processes, signatures of unexpectedly long-lived coherences have emerged in 2D ensemble spectra of various light-harvesting complexes^{77,78}. Experimental data⁷⁹ suggest that long-lived (~400 fs) quantum coherence renders energy transfer in photosynthetic systems robust in the presence of disorder, which is a prerequisite for efficient light harvesting. Here, again by removing ambiguity in the number of input photons, single-photon sources might be exploited to better investigate the quantum coherence properties at the few-photon level by providing defined spatial and temporal inputs into the pathway, as well as measuring the absolute (quantum) efficiency of the process, while correlated (entangled) photons might even allow the energy transfer process to be enhanced.

Future improvements

Apart from having applications in the study of fundamental biophysical and biological phenomena, SPSs could also inspire and catalyse the development of new instrumentation for biomedical research. One appealing idea is to use correlated photon sources to enhance the fundamental sensitivity of a microscope⁴⁷ or potentially the spatial resolution of a microscope by exploiting the reduced de Broglie wavelength of N -photon Fock states (see ref. 1 and its Supplementary Information). Another idea is to enhance the efficiency of two-photon excitation of fluorescence, a powerful tool in modern biological imaging^{80,81}. Here, the intrinsic spatiotemporal (energy–time) correlations of some quantum light sources⁸² (such as SPDC) could be exploited to increase the effective cross-section and efficiency of this process by many orders of magnitude^{83–85} and thereby overcome a major limitation of the technique, which currently requires large optical intensities that can lead to photodamage in light-sensitive biological samples (Fig. 2d).

Future developments of SPS, especially with respect to generation rate, quality ($g^{(2)}$) and the ability to generate defined multiphoton (Fock) states, will aid in the rapid adoption of these unconventional light sources in the bio-community.

Applications in fundamental quantum physics Introduction to foundational experiments

Although many properties of light can be explained in a semiclassical approach in which matter is quantized and light is treated classically,

single-photon experiments proved the non-classical nature of light⁸⁶. The ability to count single photons with avalanche photodiodes and the development of SPDC⁸⁷ and its application as a source of heralded single photons, as well as SPSs in general, made it possible to observe various quantum phenomena that had not been accessible to direct measurement before.

Needs and requirements

Most experiments that test the foundations of quantum mechanics with single photons do not impose any particularly stringent requirements on the source. In fact, most experiments can be done equally well with simple weak coherent pulses as with proper single-photon sources, unless one is testing explicit nonlinearities. Very often, foundational experiments are interferometric, and thus the longitudinal or transverse coherence of the source may be more relevant, for example in multipath delay interferometers or for illuminating slits, respectively. Foundational experiments may further demand that the source is bright, because, especially with post-selection and filtering, the achievable statistics may be limited by the interferometric stability of an experimental set-up.

On the detection side, good efficiency and low noise (dark counts) are always in demand. Usually, efficiency is not a big concern, but because it will never be perfect, for some cases this may leave loopholes open, as was the case for Bell’s-inequality experiments. Noise, however, can ruin the contrast of interferometric measurements, especially with strong filtering or post-selection. A particular requirement that has appeared in a class of foundational experiments is linearity. The properties of most single-photon detectors are influenced both by the instantaneous and by the average sustained count rate. The instantaneous count rate is limited by the dead time, from which almost any single-photon detector suffers. The gain mechanism of the detector must be recharged before it can amplify another photon detection event. The average count rate, however, often has an influence via the detector temperature or its supply current. Thus, for example, the efficiency of a detector may drop at higher count rates, before the effect of the dead time kicks in. Any of these effects influence the linearity of a single-photon detector, which can result in a false positive result, such as an artificial nonlinearity⁸⁸.

Achieved milestones

When experimental physicists managed to harness the creation and control of single photons, they immediately used them for testing fundamental principles, especially in quantum physics. In the Supplementary Information, we present a (necessarily) incomplete set of experiments and tests that covers the realization of *gedanken* experiments inspired by the pioneers of quantum theory. Here, we discuss the implementation of modern concepts, including the weak measurement of photon trajectories (weak measurements provide a new strategy for interrogating and probing quantum systems). We omit older experiments, which were already covered in earlier reviews (see ref. 89) and also loophole-free tests of non-locality that require the use of at least two photons (for testing Bell’s inequalities), which are out of the scope of this Review.

Sorkin interference experiments. Deviations from quantum mechanics are not expected for photons or other low-energy experiments. However, the most precise experiments testing the limits of quantum mechanics are nonetheless being done at low energy, for example in spectroscopy of atomic energy levels. Looking closely at any aspect of

quantum theory that has not been scrutinized before is a worthwhile endeavour. In this context, it is interesting to directly investigate the admissibility of generalized probabilistic theories, theories that go beyond quantum mechanics. Based on the ideas of Rafael Sorkin⁹⁰, one such theory predicts the existence of higher-order interference – that is, interference terms that are the product of three or more amplitudes – but at the single-particle level. This could, for example, happen if Born's rule were violated, if the probability of a certain outcome were not given by the absolute square of the amplitude. Such a scenario can be tested by a three-path interference experiment, a triple-slit experiment, where one forms a combination of the three-, two- and one-path probabilities to observe a particular outcome (position on the screen). This combination should then be identically zero, independent of the particular properties of the interference experiment. Several optical experiments^{91–93} put ever-better bounds on any hypothetical higher-order interference so that this direction of generalizing or modifying quantum physics seems to be closed.

Superposition of causal order, hypercomplex quantum mechanics.

There are other ways in which quantum mechanics might be extended or modified, for example quantum mechanics based on quaternions rather than complex numbers⁹⁴. Asher Peres⁹⁵ proposed two experimental tests for features of such a modification either via an unconventional phase relation between three scattering amplitudes or via the non-commutativity of quaternion phase shifts. The former has been put to a test in an experiment where the authors checked for residual phase shifts in a Sagnac interferometer, which realizes an interference between the two orders of two phase shifters⁹⁶. Within the experimental uncertainty, the experiment could not detect any non-commutativity, but more interesting is the debate about whether an optical experiment could be a decisive test for quaternion quantum mechanics in the first place^{97,98}.

Tunnelling time. The question of how much time elapses when a particle tunnels through an energy barrier was first answered for photons in 1993 (ref. 99). The experiment used the femtosecond-level time resolution of Hong–Ou–Mandel interference to compare the time it took a photon to tunnel through a multilayer mirror to the time it took the photon to travel through free space. Figure 3 shows the schematic of the experiment, in which a pair of photons is created by SPDC and sent to a beamsplitter where one photon goes freely and is used as a reference, while the other goes through the multilayer mirror, or not, depending on its position. Although the photon wavefunction was strongly attenuated, the authors⁹⁹ clearly showed that the tunneled photons must have propagated superluminally and found that the measured value of the tunnelling time agreed best with the expected group delay rather than with other proposed definitions of the tunnelling time.

Berry phase. The Berry or geometric phase arises in the adiabatic evolution of a quantum system around a closed loop. The initial and final state may then exhibit a phase difference, even though there was no dynamical phase shift. Although geometric phase shifts also occur in classical systems, the authors of ref. 100 were able to measure the Berry phase in a decidedly non-classical situation for single photons whose state was taken around the Poincaré sphere in a closed loop.

Experimental weak measurement of single-photon trajectories.

Every measurement involves the interaction of the system to be measured with the measuring system, often called the 'apparatus'. If this

interaction is weak – that is, if only a limited amount of entanglement is created between system and apparatus – one may only gain a limited amount of information about the system. At the same time, the disturbance of the system's free evolution caused by the measurement interaction is also reduced. Based on a proposal in ref. 101, the combination of weak interactions with a judicious choice of the pre- and post-measurement quantum states leads to the 'weak value' of a given observable. Because the combination of pre- and post-measurement state can act as a filter, it can reduce experimental noise and make effects measurable that would have been hidden in the noise otherwise. For details, we refer to the review¹⁰². Here, we select a very few works to discuss in detail.

Reference¹⁰³ reports an experiment during which the velocity and the position of a single photon were simultaneously measured using weak measurements. Exploiting the statistical data obtained by carrying out many runs makes it possible, when the weak coupling corresponds to a measurement of velocity and the strong measurement to a measurement of position, to reconstruct the local velocity of the photon and, after integration, the single-photon trajectory. In the experiment in ref. 103, the weak coupling was realized by letting light pass through a crystal endowed with the property of correlating the direction of the Poynting vector of the incoming light with the direction of polarization of the electric field at the output. Therefore, as has been argued elsewhere¹⁰⁴, one can also interpret the experiment classically in terms of the Poynting vector which is, in a classical approach, proportional to the local velocity. Another way of observing the wavefunction of a single photon was by the hologram of the photon using weak measurements. The idea was to record a hologram of a single photon probed by another reference photon on the basis of a different concept of quantum interference between two-photon probability amplitudes¹⁰⁵. It can be shown that the photon wavefunction approach provides a bridge between both interpretations, as it is possible to construct a local quantum Poynting vector in terms of the (complex) electric and magnetic single-photon wavefunctions. If one interprets trajectories obtained via weak measurements in the framework of the de Broglie–Bohm interpretation, the velocity that one finds in this formalism is a single-photon version of the Poynting vector. Such a velocity respects Einsteinian causality because it is bounded in norm by the speed of light, but it does not transform as a Minkowski four-vector (its components rather transform as elements of a second-order tensor). Therefore, the photon trajectory depends on the velocity of the observer, which is a subtle manifestation of single-photon non-locality^{106,107}: one can conceive situations where both a moving observer and an observer at rest will respectively measure a photon trajectory that remains at rest relative to each of them, which contradicts the principle of relativity and questions the reality of the reconstituted single-photon trajectories.

Observation of the spin Hall effect of light via weak measurements.

In what seems to be the first deliberate use of amplification by weak measurements, the authors of ref. 108 were able to convincingly observe the spin Hall effect for light¹⁰⁹. This is a transverse shift resulting from an effective spin–orbit interaction in a light beam upon refraction at an interface, or any other refractive index gradient that acts as the effective field. In their experiment¹⁰⁸, the authors were able to detect displacements as small as 0.1 nm at zero frequency without any extraordinary stabilization techniques.

Violation of Heisenberg's measurement–disturbance relationship via weak measurements.

In his 1930 book *The Physical Principles of the Quantum Theory*¹¹⁰, Werner Heisenberg used the thought experiment of

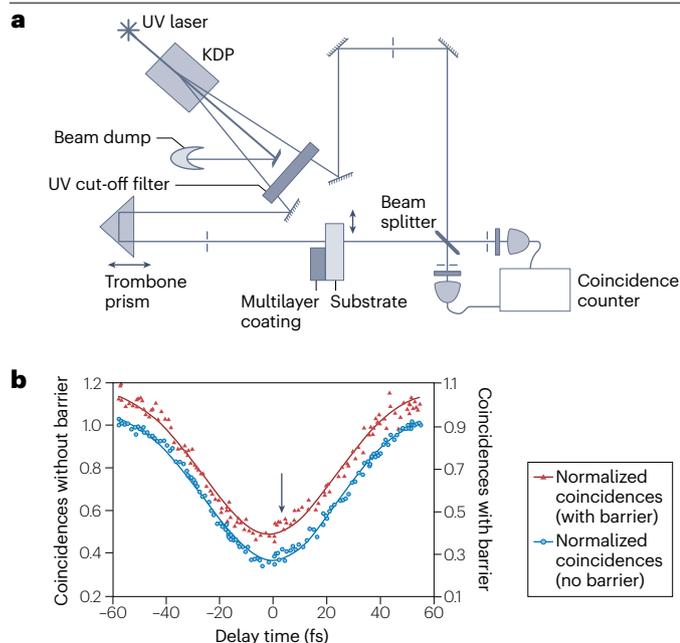


Fig. 3 | Experiment investigating quantum tunnelling. **a**, Schematic of the experiment used in ref. 99. A pair of photons is created by spontaneous parametric down-conversion and sent to a beamsplitter where one photon goes freely and is used as a reference, while the other goes through the multilayer mirror, or not, depending on its position. KDP, potassium dihydrogen phosphate; UV, ultraviolet. **b**, Experimental result with a measured delay of 1.1 fs (shown by the arrow) for the photon going through the multilayer coating mirror⁹⁹. Figure reprinted with permission from ref. 99, APS.

a microscope as one motivation for his uncertainty principle. Although the latter is an uncontested mathematical theorem, its relation to the Heisenberg microscope is tenuous at best. It has been shown that the disturbance to the quantum system under observation and the measurement uncertainty may violate the Heisenberg uncertainty inequality¹¹¹. Here, disturbance is the root-mean-square change in the prepared observable of the quantum system. The study in ref. 112 used the technique of weak measurements on single photons to reveal both the measurement uncertainty and the disturbance caused to the system, and show that the product of the two quantities violates Heisenberg's inequality^{113,114}.

Outlook

To conclude, creating, controlling and understanding the nature of a single photon remain critical and a challenge. What one can do with an SPS has evolved from *gedanken* experiments to real applications such as the ones (briefly) described above and in ref. 1.

For these applications, and probably for future ones, the following wish list is ideally necessary:

- Be able to control the photon's colour/energy: ultraviolet range for biology applications, visible range for quantum metrology and computing applications, near-infrared range for communication applications and microwave for computing and sensing.
- Achieve reproducible photon-number purity: a single photon $|1\rangle$ within a time window with requirements of more than 99% purity

in a reproducible way for most applications (quantum metrology, computing), being able to generate any N -photon Fock state $|N\rangle$ where $N > 1$.

- Produce on demand: a photon gun where a single photon or exactly N photons are generated on demand and in a controllable way would be highly desirable.
- Achieve high levels of indistinguishability: having a photon indistinguishability of high quality in a scalable way (better than 99.9% 'Hong–Ou–Mandel interference visibility').

These are the ideal conditions, but as was emphasized in this Review and in ref. 1, even though the sources are still far from perfect, they have already stimulated a variety of applications that are waiting for the perfect source to be plugged in. Furthermore, current imperfect sources are already good enough to show proof-of-principle functionality of these applications. Some applications discussed in this Review already outperform their classical counterparts, for example boson sampling.

For this wish list to be fulfilled, efforts will have to be directed to the development of new single-photon sources and the improvement of the existing ones. New materials and the control of their properties will have to be studied for more efficient and robust systems. Engineering the optical and geometrical properties of SPSs must carry on using integrated optics, cavities and photonic waveguides. On the more fundamental aspects, a deep understanding of the decoherence mechanisms and the losses will be necessary for higher-quality SPSs. Finally, optimization of resources will be required so that fewer qubits are necessary for a given quantum computing protocol or the multiplexing of channels for higher quantum key distribution rates and so on. On a more futuristic note, the exquisite properties of biological photoreceptors could serve as a starting point for developing biomimetic devices and, in this case, more efficient and advanced photon detection and emission devices. On the metrology side, a new 'quantum International System of Units' could be a source- or detector-based single-photon standard, the 'quantum candela'.

The applications of SPSs are not restricted to what has been discussed here and in ref. 1. Other applications and problems linked to single photons are emerging. In fundamental physics, for instance, a proposal was made to observe the phase shift of a single photon in an interferometer induced by gravitation¹¹⁵. Practical applications of SPSs include non-line-of-sight detection and imaging¹¹⁶ and absorption spectroscopy¹¹⁷.

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Author contributions

All authors contributed to discussing and writing the article. C.C. coordinated the contributions and provided oversight.

Competing interests

The authors declare no competing interests.

Additional information

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