photonics

MDPI

Perspective

Is Heralded Two-Photon Excited Fluorescence with Single Absorbers Possible with Current Technology?

Andreas Jechow

Community and Ecosystem Ecology, Leibniz Institute of Freshwater Ecology and Inland Fisheries (IGB), 12587 Berlin, Germany; andreas.jechow@gmx.de

Abstract: The interaction between single or a fixed number of photons with a single absorber is of fundamental interest in quantum technology. The harnessing of light matter interactions at the single particle limit has several potential applications ranging from quantum communication and quantum metrology to quantum imaging. In this perspective, a setup for heralded two-photon excited fluorescence at the single absorber level is proposed. The setup is based on a heralded two-photon source utilizing spontaneous parametric down-conversion, entanglement swapping and sum frequency generation for joint detection. This perspective aimed at triggering a discussion about the study of TPA and TPEF with only very few photons. The feasibility of the scheme is assessed by estimating the performance based on state-of-the-art technologies and losses, with the conclusion that the realization appears to be very challenging, but not completely impossible.

Keywords: multi-photon processes; quantum optics; photon statistics; fluorescence

Citation: Jechow, A. Is Heralded Two-Photon Excited Fluorescence with Single Absorbers Possible with Current Technology? *Photonics* **2022**, *9*, 52. https://doi.org/10.3390/photonics9020052

Received: 18 December 2021
Accepted: 18 January 2022
Published: 19 January 2022

1. Introduction

Maria Göppert-Mayer derived two-photon absorption (TPA) theoretically in her thesis [1] and the first experimental observation of TPA was achieved 30 years later, enabled by the invention of the laser [2]. Garrett and Kaiser observed fluorescence at 425 nm induced by light with a longer wavelength of 694 nm, marking a milestone in the field of "nonlinear optics", which for many decades was attributed to be the study of light matter interactions at high light intensities involving many photons [3]. However, with the advent of both atomic physics and quantum optics experiments involving single particles of light and matter [4–6], the field of "quantum nonlinear optics" has recently also extended towards low intensities down to the single photon level [7,8].

Two-photon microscopy (TPM) is one of the state of the art high resolution imaging technologies used in the life sciences sector [9] exploiting TPA and subsequent fluorescence, two-photon excited fluorescence (TPEF). Advantages of TPM are the deeper penetration depth into the tissue and a drastically increased depth resolution (see for example review article by Zipfel et al. [10]). TPM enabled in vivo deep brain imaging, which is of great interest in neuroscience as it made the efficient detection of beta-Amyloid plaque, a protein linked to Alzheimer's disease, possible [11]. Thus far, TPM is exclusively performed with classical light at relatively high light intensities, which is problematic because biological samples are very sensitive to optical damage [12]. Thus, reducing the optical load at the probe is paramount to prevent irreversible damage to living cells, which is done by using ultrashort pulses in TPM setups that still contain many photons.

Strategies to enhance TPA/TPEF are to utilize absorbers with an enhanced cross section, for example π-conjugated chromophores with dipolar, quadrupolar, octupolar and more extended branched structures [13] exploiting plasmonic effects on surfaces [14] or with nanoparticles [15], facilitate strong photon–absorber coupling or utilize light with tailored photon statistics [16–20].

The photon statistics of a light field is important for nonlinear processes [16] and enhanced TPA was experimentally achieved with "classical" light by utilizing thermal light instead of coherent light [19,20]. The utilization of entangled photons for TPA was proposed by Gea-Banacloche [17], as well as Javaninen and Gould [18], and later experimentally demonstrated with atoms [21] as well as molecules [22–28]. Teich and Saleh proposed to exploit entangled photons for TPM in 1997 [29] with an ideal two-photon or biphoton source. They later extended this idea towards virtual state spectroscopy [30] giving birth to the field of entangled photon or quantum spectroscopy, which has gained a lot of interest since a few years [31–35]. Entangled or correlated photons are still most commonly generated by spontaneous parametric down-conversion (SPDC) and highly efficient biphoton sources can be realized by using periodically poled (PP) materials with waveguide structures [36].

Thus far, most of the entagled TPA schemes involve rather many photons incident at the probe like bright squeezed vacuum [37], but in the context of quantum nonlinear optics the question arises: "To what extend can TPA or TPEF be observed with extremely weak fields down to the single or n-photon limit?". Heralded single photon sources based on SPDC [38] have been demonstrated and Guerreiro et al. have done seminal work on detecting weak nonlinear interactions between single photons using a heralded scheme comprising multiple nonlinear crystals [39,40].

In this perspective, a scheme for heralded TPEF is proposed utilizing parts of the approach by Guerreiro et al. [40] and exploiting other enhancement strategies for TPEF, which has potential applications in quantum microscopy and quantum spectroscopy.

2. Fundamentals and Background

2.1. Classical TPA

TPA is referred to be the "simultaneous" (It should be noted, that this is of course an unphysical term. Here, simultaneous means that the two-photons have to arrive within the lifetime of the intermediate state.) absorption of two photons by an absorber. First a photon interacts with an absorber and brings an electron from a ground state $|\psi_g\rangle$ with an energy eigenvalue of ε_g to an intermediate (nonresonant) state $|\psi_i\rangle$. This intermediate state has a very short lifetime τ_i (on the order of femtoseconds) and a complex energy eigenvalue of $\varepsilon_i + ik_i/2$. If a second photon arrives within that lifetime window, the electron can be excited to the final state $|\psi_f\rangle$ with an energy eigenvalue of ε_f . In the case of TPEF, a fluorescence photon is emitted subsequently to the absorption process (see Figure 1).

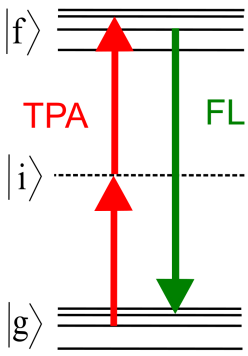

Figure 1. Sketch of two-photon excited fluorescence (TPEF). Two-photon absorption (TPA) occurs from a ground state $|\psi_g\rangle$ to a final state $|\psi_f\rangle$ via an intermediate state $|\psi_i\rangle$ with an intermediate state lifetime τ_i, followed by the subsequent emission of a fluorescence photon (FL).

The TPA rate, R_{TPA}, will depend quadratically on the intensity I or the photon flux density ϕ of the light field, when using a classical light field:

$$R_{TPA} \propto I^2 , \tag{1}$$

$$R_{TPA} = \delta_{TPA} \cdot \phi^2 , \tag{2}$$

with the classical TPA absorption cross section δ_{TPA}:

$$\delta_{TPA} = \frac{\pi}{2}\omega_1\omega_2\delta(\varepsilon_f - \varepsilon_g - \omega_1 - \omega_2) \times \left| \sum_j \left[\frac{D_{21}^{(i)}}{\Delta_1^{(i)} - i\kappa_i/2} + \frac{D_{12}^{(i)}}{\Delta_2^{(i)} - i\kappa_i/2} \right] \right|^2 , \tag{3}$$

being on the order of:

$$\delta_{TPA} \approx \sigma_{gi}\sigma_{if} \cdot \tau_i , \tag{4}$$

Here, σ_{gi} and σ_{if} are the one-photon absorption cross sections, ω_1 and ω_2 are the frequencies of the photons (or fields) driving the TPA transition, ε_f and ε_g are the energy states of the final and the ground state, respectively, $\Delta_k^{(i)} = \varepsilon_i - \varepsilon_g - \omega_k$ is an energy mismatch factor and D_{21} and D_{12} are the transition matrix elements $D_{kl}^{(i)} = \langle\psi_f|d_k|\psi_i\rangle\langle\psi_i|d_l|\psi_g\rangle$ with the electric dipole components d_k, d_l with $k, l = 1, 2$.

2.1.1. Enhanced Classical TPA

Mainly motivated to reduce photo-damage, a lot of effort has been made on the design of new absorbers with a large TPA cross section. Such chromophores can be tailored by designing donor acceptor groups, π-conjugated connectors and branching symmetry [41] including dendrimers [23]. A review on TPA chromophores with dipolar, quadrupolar, octupolar and more extended branched structures can be found in [13]. Furthermore, it was shown that by coupling fluorescent dyes to plasmonic nanostructures, such as spheres, it is possible to achieve a field enhancement resulting in an increased absorption, as well as increased fluorescence of dye molecules [42], which can be implemented on surfaces [14] or by utilizing nanoparticles [15]. Such nanoparticles can have tailored shapes and serve as nanoantennas [43,44]. Besides, enhanced absorption cross section can improve or even tailor the fluorescence by plasmonic effects [45]. The enhancement depends on the wavelength, the size of the nano-particle and also on the distance between the absorber and the fluorescent dye. In some cases, a fluorescence quenching was observed, therefore the spacer layer of the nano-particles (as sketched in Figure 2) has to be carefully adjusted depending on the dye and the excitation wavelength.

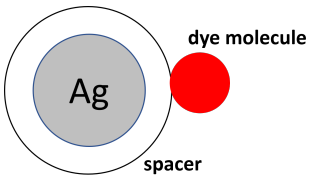

Figure 2. Sketch of a dye molecule attached to a metal nanoparticle (here silver) with a spacer layer to exploit plasmonic enhancement of absorption or fluorescence.

2.1.2. Strong Photon-Absorber Coupling

To achieve strong photon–absorber coupling, several schemes have been implemented. Most prominently, high-Q enhancement cavities have been used to ensure that the photon interaction with an absorber is enhanced [8]. However, while such cavities are excellent for fundamental tests in quantum optics, they are not suitable for many real world applications as they require extreme stability and lack scalability. For TPM applications, cavities are not well suited. Therefore, several optical interconnects for free space coupling of photons and single absorbers have been proposed and utilized, including the use of high NA lenses [46–49] or mirrors for trapped ions [50–52] or molecules [53–55]. For TPEF, it is important to not only consider the coupling between the excitation beam and the single absorber, but also to realize a high collection efficiency of fluorescence photons.

2.2. TPA and Photon Statistics

It is well known that higher order absorption processes will probe higher order correlation functions of the light fields [16,56–59]. In 1968, Mollow used pertubative theory to connect the degree of second order coherence with the TPA rate [16]. Mollow assumed, that the spectral width of the final excited energy state is large compared to the bandwidth of the field $\Delta\omega_f \gg \Delta\omega$, as typical for commonly used laser dye molecules. Mollow showed, that for weak stationary light fields the TPA rate is linearly proportional to the degree of second-order coherence function at zero time delay

$$R_{TPA}(\omega) = g^{(2)}(0)\,|D(\omega_0)|^2\,2\,\frac{\Delta\omega_f/2}{(\Delta\omega_f/2)^2 + (2\,\omega - \omega_f)^2}\,I^2\,, \tag{5}$$

with I being the intensity of the light field, ω_f being the transition frequency, $\Delta\omega_f$ being the spectral full width at half maximum of the final state and $D(\omega)$ transition.

For this work, only the influence of the photon statistic on the TPA rate is of and interest. Thus, this can be simplified to:

$$R_{TPA}(\omega) = g^{(2)}(0)\,C\,I^2\,, \tag{6}$$

with C being a constant that incorporates the properties of the absorber. When comparing the TPA rates of coherent and thermal light, one finds

$$\frac{R_{TPA}^{thermal}(\omega)}{R_{TPA}^{coherent}(\omega)} = \frac{g_{thermal}^{(2)}(0)}{g_{coherent}^{(2)}(0)} = 2\,. \tag{7}$$

In seminal work, Boitier et al. have recently demonstrated, that amplified spontaneous emission (ASE) can exhibit intensity correlations on an ultrashort timescale similar to thermal radiation [19]. They unraveled the photon bunching effect by TPA in a photomultiplier tube utilizing an interferometric setup. This was the first experimental validation of Mollows and others predictions.

Jechow et al. [20] could later demonstrate that an enhanced signal for two-photon excited fluorescence (TPEF) can be achieved when utilizing the photon statistics of the excitation light field. They used ASE from a superluminescent diode [60] in comparison to coherent light [20] and measured the fluorescence from common fluorophores and quantum dots in solution. The ASE was characterized by a thermal like photon statistics and the TPEF rate was directly proportional to the measured degree of second order coherence of the light utilized light sources. This represents a principal step towards utilizing the photon statistics in multi-photon microscopy.

2.3. Entangled TPA

Two-photon absorption with entangled photons was first experimentally demonstrated with atoms [21] and later with molecules [22–25]. Theoretical work predicted that TPA with entangled photon pairs at low flux rates will show a linear intensity dependence (see Figure 3), contrary to classical light [17,18], which was proven experimentally by Dayan et al. using broadband biphotons [61].

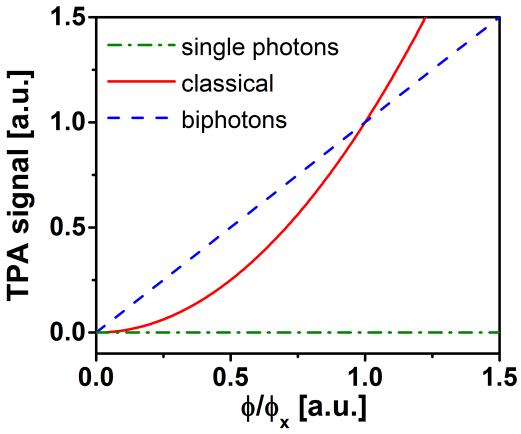

Figure 3. TPA rate R_{TPA} as a function of photon flux Φ normalized to the critical photon flux Φ_x for single photons, biphotons and classical light.

Subsequent theoretical work by Dayan [62] generalized for two-photon processes showed that below a critical flux of $\Phi_x \approx \Delta_{SPDC}$, with Δ_{SPDC} being the bandwidth of the down-converted field, the linear part will dominate, while above Φ_x the quadratic, random term will dominate. The rate enhancement is [62]:

$$\frac{R_{TPA,correlated}}{R_{TPA,uncorrelated}} = \frac{\Delta_{SPDC}}{\Delta_{TPA}} \cdot \frac{n^2 - n}{n} \cdot \frac{P_e(\tau_i - \tau_s)}{g^{(2)}_{pump}(0)}. \quad (8)$$

With n being the photons per mode ($n = 1$ at Φ_x), Δ_{TPA} being the bandwidth of the two-photon process, $g^{(2)}_{pump}(0)$ the degree of second order coherence of the light field pumping the SPDC and P_e being a normalized term describing a possible delay between signal and idler. Thus, at low excitation intensities (or small values of n), a drastic enhancement with nonclassical light is expected. Using a single photon source will result in no TPA signal.

Teich and Saleh proposed to exploit entangled photons for two-photon microscopy in 1997 [29] and later extended this idea towards virtual state spectroscopy [30]. Since a few years, several novel theoretical work has spawned in the context of entangled photon spectroscopy [31–33]. The recent tutorial by Schlawin is highly recommended for further reading [34].

3. The Proposed Setup for Heralded TPEF

Here, a new scheme for heralded TPA/TPEF experiments, exploiting several of the aforementioned schemes altogether, is proposed. The proposed system is depicted in Figure 4 and should comprise:

- A heralded two-photon scheme utilizing PP waveguide crystals,
- A high coupling between absorber and excitation field and an efficient photon collector for the fluorescence,
- A single absorber with a plasmonic enhancement antenna, and
- State-of-the-art single-photon counting devices.

The light source of the setup is inspired by seminal work by Guerrero et al. that targeted applications in quantum communication [40]. SPDC in two separate nonlinear PP waveguide crystals (SPDC1 and SPDC2) is pumped by a common pump laser (e.g., a frequency doubled diode laser [63] or a diode pumped solid state laser). The pump laser radiation is separated into two beams at a beam splitter (BS) and subsequently coupled into the waveguide crystals using high NA lenses (see, e.g., [64] using PP lithium niobate(PPLN)). In each of the waveguides a certain amount of photon pairs are generated independently, depending on the material constant, the phase matching conditions, the waveguide architecture (length, width, etc.) as well as the pump power. The light exiting the waveguides is collimated, and the pump fields are filtered out using bandpass

filters (F). The SPDC field will be centered around twice the pump wavelength (i.e., 1064 nm in our example).

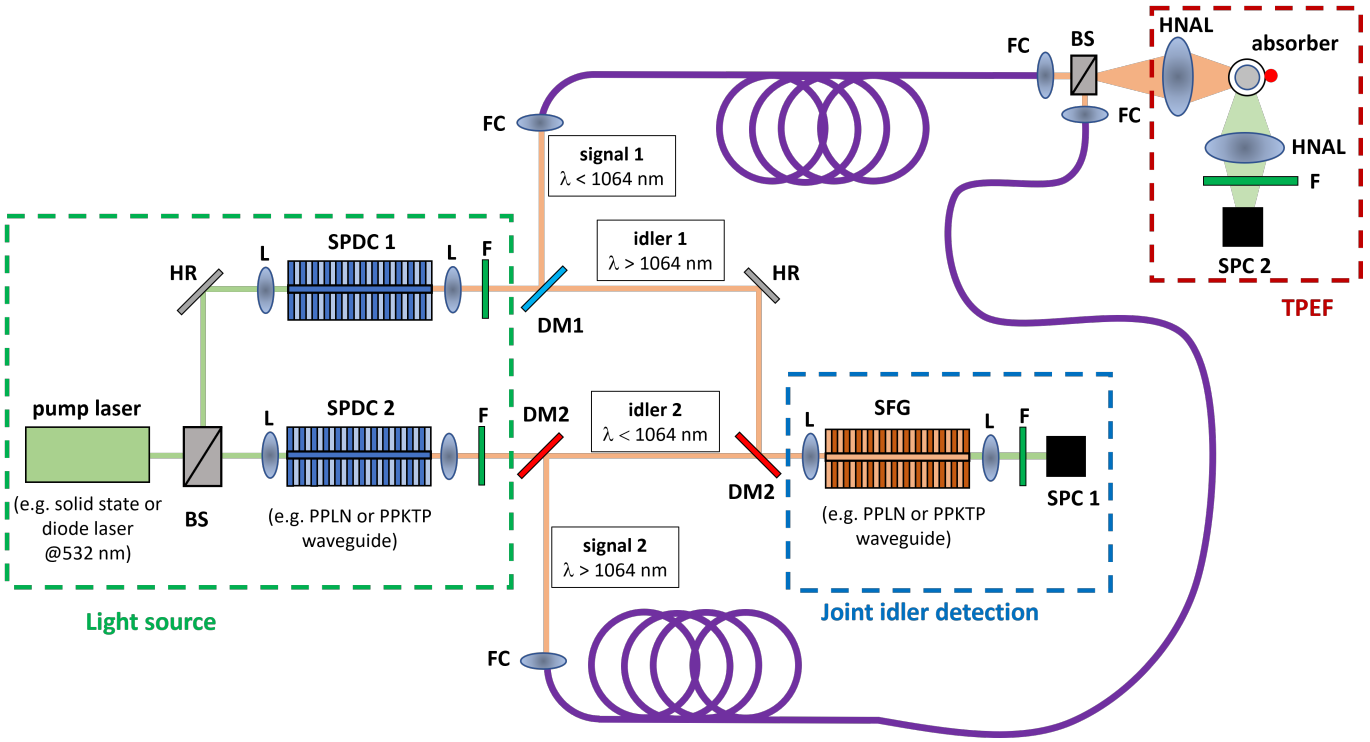

Figure 4. Sketch of a two-photon excited fluorescence (TPEF) experiment utilizing photon pairs from a heralded two-photon source to excite fluorescence in a single absorber (e.g., a dye molecule with a nano-antenna). A laser at a visible wavelength (e.g., 532 nm) is used to pump SPDC in two waveguide PP crystals (SPDC1 and SPDC2). The idler and signal photons of the SPDC sources are separated by dichroic mirrors (DM). Joint detection of the idler photons from both sources is performed in a waveguide nonlinear crystal by SFG and subsequent detection of an up-converted photon at a single photon detector (SPD 1). If two photons are jointly detected at the SFG crystal, two correlated/entangled photons must be in the two other channels. The delay between these two heralded photons can be controlled in fibers and/or by other means. The two-photons are reunified at a beam splitter (BS) and focused tightly onto the two-photon absorber using high NA optics (HNAL). The emitted fluorescence photons are collected with another high NA optic and are detected with another detector (SPD 2). The detection of the up-converted photon behind the SFG can herald the presence of two other photons that then can trigger two-photon excited fluorescence (TPEF) in a gated detection setup with an optical switch (OS) to reduce noise. BS—beam splitter, L—lens, F—optical filters, and FC—fiber coupling lenses.

In the next step, the signal and idler fields from the down-converted light fields created in each SPDC crystals are separated utilizing dichroic filters (DM1 and DM2). Subsequently, the idler field from SPDC crystal 1 (wavelengths larger than 1064 nm) and the idler field from SPDC crystal 2 (wavlengths shorter than 1064 nm) are directed towards a third periodically poled nonlinear waveguide crystal, where the two light fields are spatially overlapped and coupled into the waveguide.

Behind this waveguide crystal a single photon detector (SPC 1) is placed behind a bandpass filter. In this crystal up-conversion of the two independently generated broadband down-converted fields occurs, and a single up-converted photon is generated if two photons of the matching energy arrive at the waveguide crystal "simultaneously". Therefore, only if two photon pairs are generated "simultaneously" in the SPDC1 and SPDC2 crystals, a joint detection of the two idler photons from different SPDC crystals is possible [40]. This heralds the presence of two entangled photons in the two signal beams.

In the setup by Guerreiro et al., the two remaining outputs of the SPDC sources were each directed towards independent single photon detectors and by correlation between the three SPCs, photon triplets were detected [40] to show the possibility to herald the two photon correlation by the SFG detection. In the setup proposed here, the signal fields from SPDC crystal 1 and SPDC crystal 2 are coupled into fiber optics with an optional integrated optical switch (OS) that is opened when a photon pair is heralded at the SFG crystal. Both fiber optics require an adequate length to compensate for the slow electronics that control the shutters. At least one of the two paths requires an additional tunable optical delay line to provide fine tuning ability of any optical path length difference between the two photons. Each optical field from the fiber optics is then directed towards a beam splitter (BS), where the two light fields are spatially overlapped. (Adjusting the delay line could be realized by placing another SFG crystal at this position.) The overlapped beams are then focused tightly onto an absorber inside a lightproof box using high NA optics (HNAL) [55]. The absorber is ideally a levitating single molecule with a large TPA cross section. One way to implement the levitating single particle is to use a Paul trap architecture as shown with fluorescent dyes [65], nanoparticles [66] and proposed for large biomolecules [67]. Fluorescence of the absorber is collected by another high NA optic (HNAL) [53] that could possibly be diffractive optics [68], or a (tapered) fiber [65,69]. On the detector side, a single photon detector or a sensitive camera [20] could be used.

4. Discussion

The proposed setup could in principle be applied to study TPA/TPEF with few photons in detail. As outlined above, one major application of entangled TPEF is TPM, where the use of entangled photons (biphotons) promise a drastic signal enhancement. Another promising recent finding in using entangled photons for TPM is that Lum et al. showed that nonclassical time and energy correlations survived past 200 µm in milk and chicken tissue at room temperature, indicating that reductions in time-energy correlations are unlikely to be a limiting factor for deep tissue entangled TPA [70]. While some groups have shown large entangled TPA cross sections and enhancement factors from entanglement and even some indication for entangled TPEF with strong fields [24,25], some other groups have shown less optimistic performances and theoretical predictions [26–28]. Such enhancements have also been demonstrated recently in multiphoton processes up to the 4th order [37] using bright squeezed vacuum. While most of the other setups rely either on absorption architectures or on rather high fluxes of photon pairs, the setup presented here aims towards using very low fields.

The heralded TPEF setup outlined here is not the final leap towards TPM application, but could be utilized to test entangled two-photon absorbers at the low limit. To gain more insights at this side is important because as an enhanced signal with classical light could stem from a cascaded single-photon process and not from a real TPA process. Then, the entanglement would not provide a benefit [71]. By harnessing the absorption of only two photons in a heralded setup and by controlling the delay between the two entangled photons it appears also feasible to perform virtual state spectroscopy [30,34].

However, the question is if heralded TPEF with single absorbers is really feasible with the current technology at hand and what to expect from such a scheme in terms of performance. On the one hand, the setup relies on well known components and the heralding of photon triplets was recently demonstrated by Guerreiro et al. [40]. However, they had to integrate over an extremely large time window of 260 h to show a correlation

peak of 80 counts over a background of about 10 counts. They were limited by the dark count rate of their SPC at the SFG detection side. Their standard single photon avalanche diodes had about 3 counts per second and a response time of several 100 ps [40]. Recent progress shows highly efficient superconducting nanowire single-photon detectors are possible with time resolution below 3 ps [72], at low dark count rates below 10^{-4} per second [73] and efficiencies of 80–90% at visible wavelengths [74]. However, it has to be noted that none of the devices showed this performance at the same time.

A rough estimation of the photon flux rates gives about $\Phi_{SPDC} \approx 10^{11}$ biphotons/s assuming about 100 mW pump power at each SPDC crystal and an SPDC efficiency of 10^9 biphotons/s $\cdot$ mW as achieved in [64] with a PPLN waveguide, which is probably two orders of magnitude higher than the flux achieved by Guerreiro et al. with a pulsed solid state laser and bulk PPLN crystals [40]. Assuming about the same upconversion efficiency as Guerreiro et al. of ca. 10^{-8}, the (optimistic) SFG photon flux would be $\Phi_{SFG} \approx 10^3$ photons/s, which is much higher than the low dark count rate below 10^{-4} per second that could be achieved with the superconducting nanowire single-photon detectors [73]. Optical losses are not included yet and could be near unity with fiber optics and waveguides. Typical losses are on the order of 50–90%, giving an overall loss of 10–70%. High NA optics have shown very strong atom photon coupling efficiencies of 90% when focusing tight light fields on isolated single atoms [55] or trapped ions [49], as well as high collection efficiencies of more than 50% for fluorescence photons with a special mirror [51]. Such a laser cooled ion would also give a scattering rate of about 30% at the low intensity limit, allowing fluorescence detection at the pW level [49]. However, this would be much lower for a molecule at room temperature. Even when assuming the extremely optimistic (and most likely wrong) prediction of Saleh and Teich [29] that the entangled TPA cross section could reach that of a single photon absorption cross section at the low photon limit, one would assume several orders of magnitude lowered fluorescence photon flux. Estimating the losses of the optics to be 10^{-1} and taking the SFG photon flux of $\Phi_{SFG} \approx 10^3$ photons/s and the low dark count rate of 10^{-4}, the efficiency of the entangled TPA would need to be better than 10^{-6}. This may sound doable, but taking into account the demanding precision for the temporal "simultaneous" arrival of the entangled photons and the uncertainty for the TPA cross section enhancement, one might just end up in the noise floor.

5. Conclusions

In this perspective, the feasibility of heralded TPEF with single absorbers is discussed based on a proposed scheme extended from a setup that targets application in quantum communication [40]. Other building blocks include high NA optics and fluorescent collection schemes from single atom experiments [51,55] and enhancement by plasmonic nano-antennas [43]. Although the individual building blocks have not been tested altogether, there seems to be no obvious reason to rule out their combined functioning. However, despite utilizing current technologies, the implementation remains very challenging. One obstacle is the detection efficiency and dark counts of the detectors, which might be solvable with cryogenic superconducting nanowire single-photon detectors, which have very low dark count rates [73]. The other uncertainty lies in the efficiency of the TPA process itself. If enhancements are less than the optimistic predictions, the signal might be too weak. This perspective aimed at triggering a discussion about the study of TPA and TPEF with only very few photons. In conclusion, the complexity of the setup and the expected low signal prevent an immediate real-world application in TPM, but the setup might be a good tool for fundamental studies in the context of quantum microscopy and quantum spectroscopy [34].

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The author would like to thank Ralf Menzel, Axel Heuer, Henning Kurzke and Jan Kiethe for helpful discussions in the framework of entangled two-photon absorption.

Conflicts of Interest: The author declares no conflict of interest.

Appendix A. Photon Statistics and Nonclassical Light Sources

Appendix A.1. Photon Bunching

Different light sources emit light with different photon statistics. Generally, classical light will have a rather random temporal distribution of photons (Figure A1 middle graph). On the one hand, coherent light obeys Poissonian counting statistics on the other hand thermal light obeys Bose Einstein counting statistics. The degree of second-order coherence of a light field at a time delay τ with respect to a time t is a measure of the two-photon correlation and is defined as:

$$g^{(2)}(t,\tau) \quad = \quad \frac{\langle E^+(t)\, E^+(t+\tau)\, E^-(t+\tau)\, E^-(t)\rangle}{\langle E^+(t)\, E^-(t)\rangle^2}, \tag{A1}$$

with E^+ being the positive and E^- being negative frequency parts of the field and $\langle\rangle$ being the quantum expectations. For a perfectly coherent single-mode light field of constant amplitude the degree of second-order coherence will always be one:

$$g^{(2)}_{coherent}(t,\tau) = g^{(2)}_{coherent}(0) = 1\,. \tag{A2}$$

For a thermal (or chaotic) light field, the degree of second-order coherence will reach a value of two at zero time delay and will drop to lower values for larger time delays:

$$g^{(2)}_{thermal}(t,\tau) < g^{(2)}_{thermal}(0) = 2\,. \tag{A3}$$

Thus, the probability of detecting two photons at zero time delay with thermal light is twice as high as for coherent light. More than 60 years ago, and even before the laser was invented, Hanbury Brown and Twiss famously experimentally observed this so-called "photon bunching" [75]. However, at large time delays τ, the degree of second-order coherence for the thermal and coherent light field will become equal, both reaching values of one

$$g^{(2)}_{coherent}(t,\tau) = g^{(2)}_{thermal}(t,\tau) = 1\,. \tag{A4}$$

As typical coherence times of thermal like radiation are on the order of femtoseconds, it took more than 50 years until photon bunching on such ultra-fast timescales was observed due to limited detector bandwidths. In 2009, Boitier et al. exploited two-photon absorption in a semiconductor in combination with an interferometric auto-correlation measurements to unravel photon bunching with femtosecond temporal resolution [19].

Quantum light (or "nonclassical" light) can have a tailored photon statistics. Today, nonclassical light sources are widely used in quantum optics, especially for quantum cryptography, quantum computing and quantum communication.

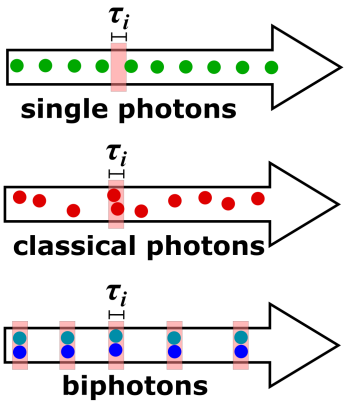

Figure A1. Illustration of the correlation of the emission from different light sources with respect to a time window τ_i. Upper graph: perfect single photon source with no two-photon correlations, middle graph: classical light source with random two-photon correlation and lower graph: perfect two-photon source that emits photon pairs or "biphotons" with perfect two-photon correlations.

Appendix A.2. Photon Pair Sources

An ideal two-photon source always emits two photons at a time (see Figure A1 lower graph) and the probability to find two photons at a time will be maximized, at least for low photon flux densities. Entangled photon pairs or "biphotons" can have such strong spatio-temporal correlations [76,77] and have opened up many key findings in the field of quantum optics. They are most commonly generated by spontaneous parametric down conversion (SPDC) in a nonlinear optical crystal [78–80]. In SPDC, one pump photon with a high energy will decay into two photons of lower energy. Historically, one photon is called "idler" and the other is called "signal". Momentum and energy conservation applies so the sum of the energies of signal and idler equals that of the original pump photon. The peculiar behavior of these biphotons have been utilized to demonstrate stunning experiments impossible with classical light, e.g., quantum teleportation [81]. Entangled photon pairs can also be obtained from semiconductors such as a quantum dot embedded in an LED [82], by four-wave-mixing [83], SPDC in semiconductors with Bragg waveguides [84] or from on chip SPDC in a laser diode with a super-lattice structure [85,86]. However, most of these systems are still under development and not mature, yet.

Appendix A.3. Single Photon Sources

An ideal single photon source (Figure A1 upper graph) always emits just one photon at a time and the chance to find two photons at a time will be zero, which is called "anti-bunching". Within the last few years, a lot of progress was made with such single photon sources [87]. Single photons can be obtained from heralded sources exploiting parametric down-conversion [38,88,89] or single emitters like single molecules [90,91], single atoms [92,93], quantum dots [94–96], silicon nanowires [97], silicon carbide [98] or nitrogen vacancies in diamond [99,100].

References

1. Göppert-Mayer, M. Über Elementarakte mit zwei Quantensprüngen. *Ann. Phys.* **1931**, *401*, 273–294. [CrossRef]
2. Kaiser, W.; Garrett, C.G.B. Two-Photon Excitation in CaF_2: Eu^{2+}. *Phys. Rev. Lett.* **1961**, *7*, 229–231. [CrossRef]
3. Boyd, R.W. *Nonlinear Optics*, 3rd ed.; Academic Press: San Diego, CA, USA; Burlington, MA, USA, 2008.
4. Aspect, A.; Grangier, P.; Roger, G. Experimental Realization of Einstein-Podolsky-Rosen-Bohm *Gedankenexperiment*: A New Violation of Bell's Inequalities. *Phys. Rev. Lett.* **1982**, *49*, 91–94. [CrossRef]
5. Polycarpou, C.; Cassemiro, K.N.; Venturi, G.; Zavatta, A.; Bellini, M. Adaptive Detection of Arbitrarily Shaped Ultrashort Quantum Light States. *Phys. Rev. Lett.* **2012**, *109*, 053602. [CrossRef]
6. Manning, A.G.; Khakimov, R.I.; Dall, R.G.; Truscott, A.G. Wheeler's delayed-choice gedanken experiment with a single atom. *Nat. Phys.* **2015**, *11*, 539–542. [CrossRef]
7. Dayan, B.; Parkins, A.S.; Aoki, T.; Ostby, E.P.; Vahala, K.J.; Kimble, H.J. A Photon Turnstile Dynamically Regulated by One Atom. *Science* **2008**, *319*, 1062–1065. [CrossRef]

8. Chang, D.E.; Vuletic, V.; Lukin, M.D. Quantum nonlinear optics [mdash] photon by photon. *Nat. Photonics* **2014**, *8*, 685–694. [CrossRef]
9. Denk, W.; Strickler, J.; Webb, W. Two-photon laser scanning fluorescence microscopy. *Science* **1990**, *248*, 73–76. [CrossRef]
10. Zipfel, W.R.; Williams, R.M.; Webb, W.W. Nonlinear magic: Multiphoton microscopy in the biosciences. *Nat. Biotech.* **2003**, *21*, 1369–1377. [CrossRef]
11. Dong, J.; Revilla-Sanchez, R.; Moss, S.; Haydon, P.G. Multiphoton in vivo imaging of amyloid in animal models of Alzheimer's disease. *Neuropharmacology* **2010**, *59*, 268–275. [CrossRef]
12. Hopt, A.; Neher, E. Highly nonlinear photodamage in two-photon fluorescence microscopy. *Biophys. J.* **2001**, *80*, 2029–2036. [CrossRef]
13. Terenziani, F.; Katan, C.; Badaeva, E.; Tretiak, S.; Blanchard-Desce, M. Enhanced Two-Photon Absorption of Organic Chromophores: Theoretical and Experimental Assessments. *Adv. Mater.* **2008**, *20*, 4641–4678. [CrossRef]
14. Wenseleers, W.; Stellacci, F.; Meyer-Friedrichsen, T.; Mangel, T.; Bauer, C.A.; Pond, S.J.; Marder, S.R.; Perry, J.W. Five orders-of-magnitude enhancement of two-photon absorption for dyes on silver nanoparticle fractal clusters. *J. Phys. Chem. B* **2002**, *106*, 6853–6863. [CrossRef]
15. Cohanoschi, I.; Yao, S.; Belfield, K.D.; Hernández, F.E. Effect of the concentration of organic dyes on their surface plasmon enhanced two-photon absorption cross section using activated Au nanoparticles. *J. Appl. Phys.* **2007**, *101*, 086112. [CrossRef]
16. Mollow, B.R. Two-Photon Absorption and Field Correlation Functions. *Phys. Rev.* **1968**, *175*, 1555–1563. [CrossRef]
17. Gea-Banacloche, J. Two-photon absorption of nonclassical light. *Phys. Rev. Lett.* **1989**, *62*, 1603–1606. [CrossRef]
18. Javanainen, J.; Gould, P.L. Linear intensity dependence of a two-photon transition rate. *Phys. Rev. A* **1990**, *41*, 5088–5091. [CrossRef]
19. Boitier, F.; Godard, A.; Rosencher, E.; Fabre, C. Measuring photon bunching at ultrashort timescale by two-photon absorption in semiconductors. *Nat. Phys.* **2009**, *5*, 267–270. [CrossRef]
20. Jechow, A.; Seefeldt, M.; Kurzke, H.; Heuer, A.; Menzel, R. Enhanced two-photon excited fluorescence from imaging agents using true thermal ligh. *Nat. Photonics* **2013**, *7*, 973. [CrossRef]
21. Georgiades, N.P.; Polzik, E.S.; Edamatsu, K.; Kimble, H.J.; Parkins, A.S. Nonclassical Excitation for Atoms in a Squeezed Vacuum. *Phys. Rev. Lett.* **1995**, *75*, 3426–3429. [CrossRef] [PubMed]
22. Lee, D.I.; Goodson, T., III. Entangled Photon Absorption in an Organic Porphyrin Dendrimer. *J. Phys. Chem. B* **2006**, *110*, 25582–25585. [CrossRef] [PubMed]
23. Harpham, M.R.; Süzer, Ö.; Ma, C.Q.; Bäuerle, P.; Goodson, T., III. Thiophene Dendrimers as Entangled Photon Sensor Materials. *J. Am. Chem. Soc.* **2009**, *131*, 973–979. [CrossRef]
24. Upton, L.; Harpham, M.; Süzer, Ö.; Richter, M.; Mukamel, S.; Goodson, T., III. Optically Excited Entangled States in Organic Molecules Illuminate the Dark. *J. Phys. Chem. Lett.* **2013**, *4*, 2046–2052. [CrossRef] [PubMed]
25. Villabona-Monsalve, J.P.; Calderón-Losada, O.; Nuñez Portela, M.; Valencia, A. Entangled Two Photon Absorption Cross Section on the 808 nm Region for the Common Dyes Zinc Tetraphenylporphyrin and Rhodamine B. *J. Phys. Chem. A* **2017**, *121*, 7869–7875. [CrossRef] [PubMed]
26. Parzuchowski, K.M.; Mikhaylov, A.; Mazurek, M.D.; Wilson, R.N.; Lum, D.J.; Gerrits, T.; Camp, C.H., Jr.; Stevens, M.J.; Jimenez, R. Setting Bounds on Entangled Two-Photon Absorption Cross Sections in Common Fluorophores. *Phys. Rev. Appl.* **2021**, *15*, 044012. [CrossRef]
27. Landes, T.; Raymer, M.G.; Allgaier, M.; Merkouche, S.; Smith, B.J.; Marcus, A.H. Quantifying the enhancement of two-photon absorption due to spectral-temporal entanglement. *Opt. Express* **2021**, *29*, 20022–20033. [CrossRef] [PubMed]
28. Corona-Aquino, S.; Calderón-Losada, O.; Li-Gómez, M.Y.; Cruz-Ramirez, H.; Alvarez-Venicio, V.; Carreón-Castro, M.d.P.; León-Montiel, R.d.J.; U'Ren, A.B. Experimental study on the effects of photon-pair temporal correlations in entangled two-photon absorption. *arXiv* **2021**, arXiv:2101.10987.
29. Teich, M.C.; Saleh, B.E. Entangled-photon microscopy. *Cesk. Cas. Fyz* **1997**, *47*, 3–8.
30. Saleh, B.E.; Jost, B.M.; Fei, H.B.; Teich, M.C. Entangled-photon virtual-state spectroscopy. *Phys. Rev. Lett.* **1998**, *80*, 3483. [CrossRef]
31. Schlawin, F.; Dorfman, K.E.; Fingerhut, B.P.; Mukamel, S. Manipulation of two-photon-induced fluorescence spectra of chromophore aggregates with entangled photons: A simulation study. *Phys. Rev. A* **2012**, *86*, 023851. [CrossRef]
32. Schlawin, F.; Dorfman, K.E.; Fingerhut, B.P.; Mukamel, S. Suppression of population transport and control of exciton distributions by entangled photons. *Nat. Commun.* **2013**, *4*, 1782. [CrossRef] [PubMed]
33. Dorfman, K.E.; Schlawin, F.; Mukamel, S. Nonlinear optical signals and spectroscopy with quantum light. *Rev. Mod. Phys.* **2016**, *88*, 045008. [CrossRef]
34. Schlawin, F. Entangled photon spectroscopy. *J. Phys. At. Mol. Opt. Phys.* **2017**, *50*, 203001. [CrossRef]
35. Mertenskötter, L.; Busch, K.; León-Montiel, R.d.J. Entangled two-photon absorption spectroscopy with varying pump wavelengths. *JOSA B* **2021**, *38*, C63–C68. [CrossRef]
36. Jechow, A.; Lichtner, M.; Menzel, R.; Radziunas, M.; Skoczowsky, D.; Vladimirov, A.G. Stripe-array diode-laser in an off-axis external cavity: Theory and experiment. *Opt. Express* **2009**, *17*, 19599–19604. [CrossRef] [PubMed]
37. Spasibko, K.Y.; Kopylov, D.A.; Krutyanskiy, V.L.; Murzina, T.V.; Leuchs, G.; Chekhova, M.V. Multiphoton Effects Enhanced due to Ultrafast Photon-Number Fluctuations. *Phys. Rev. Lett.* **2017**, *119*, 223603. [CrossRef]

38. U'Ren, A.B.; Silberhorn, C.; Erdmann, R.; Banaszek, K.; Grice, W.P.; Walmsley, I.A.; Raymer, M.G. Generation of Pure-State Single-Photon Wavepackets by Conditional Preparation Based on Spontaneous Parametric Downconversion. *Las. Phys.* **2005**, *15*, 146–161.
39. Guerreiro, T.; Pomarico, E.; Sanguinetti, B.; Sangouard, N.; Pelc, J.S.; Langrock, C.; Fejer, M.M.; Zbinden, H.; Thew, R.T.; Gisin, N. Interaction of independent single photons based on integrated nonlinear optics. *Nat. Commun.* **2013**, *4*, 2324. [CrossRef]
40. Guerreiro, T.; Martin, A.; Sanguinetti, B.; Pelc, J.S.; Langrock, C.; Fejer, M.M.; Gisin, N.; Zbinden, H.; Sangouard, N.; Thew, R.T. Nonlinear Interaction between Single Photons. *Phys. Rev. Lett.* **2014**, *113*, 173601. [CrossRef] [PubMed]
41. Albota, M.; Beljonne, D.; Brédas, J.L.; Ehrlich, J.E.; Fu, J.Y.; Heikal, A.A.; Hess, S.E.; Kogej, T.; Levin, M.D.; Marder, S.R.; et al. Design of Organic Molecules with Large Two-Photon Absorption Cross Sections. *Science* **1998**, *281*, 1653–1656. [CrossRef]
42. Kano, H.; Kawata, S. Two-photon-excited fluorescence enhanced by a surface plasmon. *Opt. Lett.* **1996**, *21*, 1848–1850. [CrossRef] [PubMed]
43. Sivapalan, S.T.; Vella, J.H.; Yang, T.K.; Dalton, M.J.; Swiger, R.N.; Haley, J.E.; Cooper, T.M.; Urbas, A.M.; Tan, L.S.; Murphy, C.J. Plasmonic Enhancement of the Two Photon Absorption Cross Section of an Organic Chromophore Using Polyelectrolyte-Coated Gold Nanorods. *Langmuir* **2012**, *28*, 9147–9154. [CrossRef] [PubMed]
44. Kühn, S.; Håkanson, U.; Rogobete, L.; Sandoghdar, V. Enhancement of Single-Molecule Fluorescence Using a Gold Nanoparticle as an Optical Nanoantenna. *Phys. Rev. Lett.* **2006**, *97*, 017402. [CrossRef] [PubMed]
45. Lakowicz, J.R. Plasmonics in biology and plasmon-controlled fluorescence. *Plasmonics* **2006**, *1*, 5–33. [CrossRef]
46. Streed, E.W.; Norton, B.G.; Jechow, A.; Weinhold, T.J.; Kielpinski, D. Imaging of trapped ions with a microfabricated optic for quantum information processing. *Phys. Rev. Lett.* **2011**, *106*, 010502. [CrossRef] [PubMed]
47. Jechow, A.; Norton, B.; Haendel, S.; Blūms, V.; Streed, E.; Kielpinski, D. Controllable optical phase shift over one radian from a single isolated atom. *Phys. Rev. Lett.* **2013**, *110*, 113605. [CrossRef]
48. Jechow, A.; Streed, E.W.; Norton, B.G.; Petrasiunas, M.J.; Kielpinski, D. Wavelength-scale imaging of trapped ions using a phase Fresnel lens. *Opt. Lett.* **2011**, *36*, 1371–1373. [CrossRef]
49. Streed, E.W.; Jechow, A.; Norton, B.G.; Kielpinski, D. Absorption imaging of a single atom. *Nat. Commun.* **2012**, *3*, 933. [CrossRef]
50. Shu, G.; Kurz, N.; Dietrich, M.; Blinov, B. Efficient fluorescence collection from trapped ions with an integrated spherical mirror. *Phys. Rev. A* **2010**, *81*, 042321. [CrossRef]
51. Maiwald, R.; Golla, A.; Fischer, M.; Bader, M.; Heugel, S.; Chalopin, B.; Sondermann, M.; Leuchs, G. Collecting more than half the fluorescence photons from a single ion. *Phys. Rev. A* **2012**, *86*, 043431. [CrossRef]
52. Ghadimi, M.; Blūms, V.; Norton, B.G.; Fisher, P.M.; Connell, S.C.; Amini, J.M.; Volin, C.; Hayden, H.; Pai, C.S.; Kielpinski, D.; et al. Scalable ion–photon quantum interface based on integrated diffractive mirrors. *NPJ Quantum Inf.* **2017**, *3*, 4. [CrossRef]
53. Celebrano, M.; Kukura, P.; Renn, A.; Sandoghdar, V. Single-molecule imaging by optical absorption. *Nat. Phot.* **2011**, *5*, 95–98. [CrossRef]
54. Tey, M.K.; Chen, Z.; Aljunid, S.A.; Chng, B.; Maslennikov, G.; Kurtsiefer, C. Strong interaction between light and a single trapped atom without the need for a cavity. *Nat. Phys.* **2008**, *4*, 924–927. [CrossRef]
55. Tey, M.K.; Maslennikov, G.; Liew, T.C.H.; Aljunid, S.A.; Huber, F.; Chng, B.; Chen, Z.; Scarani, V.; Kurtsiefer, C. Interfacing light and single atoms with a lens. *New J. Phys* **2009**, *11*, 043011. [CrossRef]
56. Teich, M.C.; Wolga, G.J. Multiple-Photon Processes and Higher Order Correlation Functions. *Phys. Rev. Lett.* **1966**, *16*, 625–628. [CrossRef]
57. Lambropoulos, P.; Kikuchi, C.; Osborn, R.K. Coherence and Two-Photon Absorption. *Phys. Rev.* **1966**, *144*, 1081–1086. [CrossRef]
58. Shen, Y.R. Quantum Statistics of Nonlinear Optics. *Phys. Rev.* **1967**, *155*, 921–931. [CrossRef]
59. Agarwal, G.S. Field-Correlation Effects in Multiphoton Absorption Processes. *Phys. Rev. A* **1970**, *1*, 1445–1459. [CrossRef]
60. Jechow, A.; Raab, V.; Menzel, R. Tunable 6.8 W narrow bandwidth emission from a single-stripe continuous-wave broad-area laser diode in a simple external cavity. *Appl. Opt.* **2008**, *47*, 1447–1450. [CrossRef]
61. Dayan, B.; Pe'er, A.; Friesem, A.A.; Silberberg, Y. Nonlinear Interactions with an Ultrahigh Flux of Broadband Entangled Photons. *Phys. Rev. Lett.* **2005**, *94*, 043602. [CrossRef] [PubMed]
62. Dayan, B. Theory of two-photon interactions with broadband down-converted light and entangled photons. *Phys. Rev. A* **2007**, *76*, 043813. [CrossRef]
63. Jechow, A.; Menzel, R.; Paschke, K.; Erbert, G. Blue-green light generation using high brilliance edge emitting diode lasers. *Laser Photonics Rev.* **2010**, *4*, 633–655. [CrossRef]
64. Jechow, A.; Heuer, A.; Menzel, R. High brightness, tunable biphoton source at 976 nm for quantum spectroscopy. *Opt. Express* **2008**, *16*, 13439–13449. [CrossRef] [PubMed]
65. Gregor, M.; Kuhlicke, A.; Benson, O. Soft-landing and optical characterization of a preselected single fluorescent particle on a tapered optical fiber. *Opt. Express* **2009**, *17*, 24234–24243. [CrossRef]
66. Alda, I.; Berthelot, J.; Rica, R.A.; Quidant, R. Trapping and manipulation of individual nanoparticles in a planar Paul trap. *Appl. Phys. Lett.* **2016**, *109*, 163105. [CrossRef]
67. Streed, E.W. Unfolding Large Biomolecules. *arXiv* **2012**, arXiv:1211.3168.
68. Norton, B.; Streed, E.; Petrasiunas, M.; Jechow, A.; Kielpinski, D. Millikelvin spatial thermometry of trapped ions. *New J. Phys.* **2011**, *13*, 113022. [CrossRef]

69. VanDevender, A.P.; Colombe, Y.; Amini, J.; Leibfried, D.; Wineland, D.J. Efficient Fiber Optic Detection of Trapped Ion Fluorescence. *Phys. Rev. Lett.* **2010**, *105*, 023001. [CrossRef] [PubMed]
70. Lum, D.J.; Mazurek, M.D.; Mikhaylov, A.; Parzuchowski, K.M.; Wilson, R.N.; Jimenez, R.; Gerrits, T.; Stevens, M.J.; Cicerone, M.T.; Camp, C.H. Witnessing the survival of time-energy entanglement through biological tissue and scattering media. *Biomed. Opt. Express* **2021**, *12*, 3658–3670. [CrossRef]
71. Stefanov, A. On the role of entanglement in two-photon metrology. *Quantum Sci. Technol.* **2017**, *2*, 025004. [CrossRef]
72. Korzh, B.; Zhao, Q.Y.; Allmaras, J.P.; Frasca, S.; Autry, T.M.; Bersin, E.A.; Beyer, A.D.; Briggs, R.M.; Bumble, B.; Colangelo, M.; et al. Demonstration of sub-3 ps temporal resolution with a superconducting nanowire single-photon detector. *Nat. Photonics* **2020**, *14*, 250–255. [CrossRef]
73. Shibata, H.; Shimizu, K.; Takesue, H.; Tokura, Y. Ultimate low system dark-count rate for superconducting nanowire single-photon detector. *Opt. Lett.* **2015**, *40*, 3428–3431. [CrossRef]
74. Esmaeil Zadeh, I.; Los, J.W.; Gourgues, R.B.; Chang, J.; Elshaari, A.W.; Zichi, J.R.; Van Staaden, Y.J.; Swens, J.P.; Kalhor, N.; Guardiani, A.; et al. Efficient single-photon detection with 7.7 ps time resolution for photon-correlation measurements. *ACS Photonics* **2020**, *7*, 1780–1787. [CrossRef]
75. Hanbury Brown, R.; Twiss, R.Q. Correlation between Photons in two Coherent Beams of Light. *Nature* **1956**, *177*, 27–29. [CrossRef]
76. Burnham, D.C.; Weinberg, D.L. Observation of Simultaneity in Parametric Production of Optical Photon Pairs. *Phys. Rev. Lett.* **1970**, *25*, 84–87. [CrossRef]
77. Jost, B.; Sergienko, A.; Abouraddy, A.; Saleh, B.; Teich, M. Spatial correlations of spontaneously down-converted photon pairs detected with asingle-photon-sensitive CCD camera. *Opt. Express* **1998**, *3*, 81–88. [CrossRef] [PubMed]
78. Harris, S.E.; Oshman, M.K.; Byer, R.L. Observation of Tunable Optical Parametric Fluorescence. *Phys. Rev. Lett.* **1967**, *18*, 732–734. [CrossRef]
79. Malygin, A.A.; Perin, A.N.; Sergienko, A.V. Efficient Generator of a Two-Photon field of visible radiation. *Sov. J. Quantum Electron.* **1981**, *11*, 939. [CrossRef]
80. Kwiat, P.G.; Mattle, K.; Weinfurter, H.; Zeilinger, A.; Sergienko, A.V.; Shih, Y. New High-Intensity Source of Polarization-Entangled Photon Pairs. *Phys. Rev. Lett.* **1995**, *75*, 4337–4341. [CrossRef] [PubMed]
81. Bouwmeester, D.; Pan, J.W.; Mattle, K.; Eibl, M.; Weinfurter, H.; Zeilinger, A. Experimental quantum teleportation. *Nature* **1997**, *390*, 575–579. [CrossRef]
82. Salter, C.L.; Stevenson, R.M.; Farrer, I.; Nicoll, C.A.; Ritchie, D.A.; Shields, A.J. An entangled-light-emitting diode. *Nature* **2010**, *465*, 594–597. [CrossRef]
83. Wang, L.J.; Hong, C.K.; Friberg, S.R. Generation of correlated photons via four-wave mixing in optical fibres. *J. Opt. B Quantum Semiclassical Opt.* **2001**, *3*, 346. [CrossRef]
84. Vallés, A.; Hendrych, M.; Svozilík, J.; Machulka, R.; Abolghasem, P.; Kang, D.; Bijlani, B.J.; Helmy, A.S.; Torres, J.P. Generation of polarization-entangled photon pairs in a Bragg reflection waveguide. *Opt. Express* **2013**, *21*, 10841–10849. [CrossRef] [PubMed]
85. Boitier, F.; Orieux, A.; Autebert, C.; Lemaître, A.; Galopin, E.; Manquest, C.; Sirtori, C.; Favero, I.; Leo, G.; Ducci, S. Electrically Injected Photon-Pair Source at Room Temperature. *Phys. Rev. Lett.* **2014**, *112*, 183901. [CrossRef] [PubMed]
86. Autebert, C.; Maltese, G.; Halioua, Y.; Boitier, F.; Lemaître, A.; Amanti, M.; Sirtori, C.; Ducci, S. Electrically Injected Twin Photon Emitting Lasers at Room Temperature. *Technologies* **2016**, *4*, 24. [CrossRef]
87. Lounis, B.; Orrit, M. Single-photon sources. *Rep. Prog. Phys.* **2005**, *68*, 1129. [CrossRef]
88. Mosley, P.J.; Lundeen, J.S.; Smith, B.J.; Wasylczyk, P.; U'Ren, A.B.; Silberhorn, C.; Walmsley, I.A. Heralded Generation of Ultrafast Single Photons in Pure Quantum States. *Phys. Rev. Lett.* **2008**, *100*, 133601. [CrossRef]
89. Levine, Z.H.; Fan, J.; Chen, J.; Ling, A.; Migdall, A. Heralded, pure-state single-photon source based on a Potassium Titanyl Phosphate waveguide. *Opt. Express* **2010**, *18*, 3708–3718. [CrossRef]
90. Wrigge, G.; Gerhardt, I.; Hwang, J.; Zumofen, G.; Sandoghdar, V. Efficient coupling of photons to a single molecule and the observation of its resonance fluorescence. *Nat. Phys.* **2008**, *4*, 60–66. [CrossRef]
91. Lee, K.G.; Chen, X.W.; Eghlidi, H.; Kukura, P.; Lettow, R.; Renn, A.; Sandoghdar, V.; Gotzinger, S. A planar dielectric antenna for directional single-photon emission and near-unity collection efficiency. *Nat. Photonics* **2011**, *5*, 166–169. [CrossRef]
92. Keller, M.; Lange, B.; Hayasaka, K.; Lange, W.; Walther, H. Continuous generation of single photons with controlled waveform in an ion-trap cavity system. *Nature* **2004**, *431*, 1075–1078. [CrossRef] [PubMed]
93. McKeever, J.; Boca, A.; Boozer, A.D.; Miller, R.; Buck, J.R.; Kuzmich, A.; Kimble, H.J. Deterministic Generation of Single Photons from One Atom Trapped in a Cavity. *Science* **2004**, *303*, 1992–1994. [CrossRef]
94. Michler, P.; Imamoglu, A.; Mason, M.D.; Carson, P.J.; Strouse, G.F.; Buratto, S.K. Quantum correlation among photons from a single quantum dot at room temperature. *Nature* **2000**, *406*, 968–970. [CrossRef]
95. Kuhlmann, A.V.; Prechtel, J.H.; Houel, J.; Ludwig, A.; Reuter, D.; Wieck, A.D.; Warburton, R.J. Transform-limited single photons from a single quantum dot. *Nat. Commun.* **2015**, *6*, 8204. [CrossRef] [PubMed]
96. Schnauber, P.; Thoma, A.; Heine, C.V.; Schlehahn, A.; Gantz, L.; Gschrey, M.; Schmidt, R.; Hopfmann, C.; Wohlfeil, B.; Schulze, J.H.; et al. Bright single-photon sources based on anti-reflection coated deterministic quantum dot microlenses. *Technologies* **2015**, *4*, 1. [CrossRef]
97. Zhang, X.; Jiang, R.; Bell, B.A.; Choi, D.Y.; Chae, C.J.; Xiong, C. Interfering Heralded Single Photons from Two Separate Silicon Nanowires Pumped at Different Wavelengths. *Technologies* **2016**, *4*, 25. [CrossRef]

98. Boretti, A.; Rosa, L. Latest advances in the generation of single photons in silicon carbide. *Technologies* **2016**, *4*, 16. [CrossRef]
99. Sipahigil, A.; Goldman, M.L.; Togan, E.; Chu, Y.; Markham, M.; Twitchen, D.J.; Zibrov, A.S.; Kubanek, A.; Lukin, M.D. Quantum Interference of Single Photons from Remote Nitrogen-Vacancy Centers in Diamond. *Phys. Rev. Lett.* **2012**, *108*, 143601. [CrossRef]
100. Schietinger, S.; Barth, M.; Aichele, T.; Benson, O. Plasmon-Enhanced Single Photon Emission from a Nanoassembled Metal-Diamond Hybrid Structure at Room Temperature. *Nano Lett.* **2009**, *9*, 1694–1698. [CrossRef] [PubMed]