Quantum Sensing with Extreme Light

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Optical nonlinear conversion processes are ubiquitously applied to scientific as well as industrial tasks. In particular, nonlinear processes are employed to generate radiation in many frequency ranges. In plenty of these nonlinear processes, the generation of paired photons occurs — the so-called signal and idler photons. Although this type of generation has undergone a tremendous development over the last decades, either the generated signal or the idler radiation has been used experimentally. In contrast, novel quantum-based measurement principles enable the usage of both partners of the generated photon pairs based on their correlation. These measurement approaches have an enormous potential for future applications, as they allow to transfer information from one spectral range to another. In particular, spectral ranges where photon generation and detection is particularly challenging can benefit from this principle. Above all, these include the extreme frequency ranges, such as on the low-frequency side the mid to far infrared or even the terahertz spectral range, but also on the high-frequency side the ultraviolet or X-ray spectral range. In this review article, theoretical and experimental developments based on correlated biphotons are described specifically for the extreme spectral regions.

1. Introduction

Since the discovery of the nature of visible light, the exploitation of the entire electromagnetic spectrum has revolutionized many areas of today’s life. Access to new spectral ranges always led to exciting new applications. High-energy photons (X-ray, ultraviolet [UV]) have become indispensable in medicine,[1] biology,[2] and non-destructive testing.[3] Photons with less energy (near-infrared [NIR], mid-infrared [MIR], terahertz) offer interesting applications such as gas analysis[4] and non-destructive testing[5, 6] as well. However, these frequency ranges require the use of appropriate sources and detectors.

With the invention of the laser and the possibilities of nonlinear optics, a new powerful tool has been available to generate a large variety of bright sources at various wavelengths. Nonlinear conversion (second harmonic generation) was observed for the first time half a century ago by Franken et al. shortly after the first laser was demonstrated.[7] This experiment is often taken to be the starting point of the field of nonlinear optics. Due to the nonlinear reaction of a material to the irradiation with an intense optical pump field, additional frequency components are generated. Whether these frequency components are twice the irradiated frequency (second harmonic) or add up to the irradiated frequency (spontaneous parametric down-conversion [SPDC]) depends mainly on phase matching.

In the case of SPDC the pump photons are converted in a nonlinear medium to a pair of photons whose energy sum is equivalent to the energy of the pump photon. The two photons generated are historically called signal and idler. However, already the naming shows that mainly one, the signal, was used and the other, the idler, served exclusively for the generation. Since the output of SPDC is very weak, various possibilities have been developed to amplify the process by adding one of these radiations either from the outside or in a repetitive fashion, as in the case of optical parametric amplifiers (OPA) and optical parametric oscillators (OPO). With these methods it is possible to optically produce intense radiation in spectral ranges that were difficult to reach before. Therefore, these methods are used until today.[8–12]

But now — a century after discovering the quantum nature of light — these quantum properties are still opening up new exciting optical concepts. In particular, the use of entangled or correlated photons allows to access spectral regions, which are classically difficult to address, either due to the lack of appropriate sources or more commonly due to the lack of detectors. In the 1990s, the idea of not only using the generated signal or idler photons from nonlinear generation, but the correlation between them emerged. In nonlinear interferometers, one of the generated beams interacts with the sample — called idler beam in the
following — while the other one — the signal beam — does not. If the idler and the signal beam of such a nonlinear source are superimposed with the respective beams of a second source, interference occurs. While the idler photons can be dumped, the interference of the signal photons also carries information about the properties of the sample, even though none of the signal photons interacted with it. The interference of these quantum-based interferometers is caused by the superposition of the two possible biphoton states which leads to the loss of which-path information. Much like the double-slit experiment this effect also occurs when the system is pumped by single photons.[13]

This concept is especially useful due to two reasons. First, it provides the ability to measure with alternative photon detector devices — devices which are by themselves not suitable for the addressed spectral range. This is of utmost interest for spectral regions such as the microwave or terahertz region. Due to widespread consumer interest, silicon-based sensing technology is most advanced in the visible range, making it most sensitive and cost effective.[14,15] Second, this measurement principle allows for minimized photon dose interacting with the samples,[16] which is crucial for the investigation of biological samples with UV or X-ray photons. These two aspects underline the perspective benefits, to catch up with and possibly even surpass classical measurement concepts.

While the theoretical concept was already proposed about 3 decades ago[17] and first demonstration experiments were carried out in the following years,[18–20] the principle is now transferred to new spectral ranges with more than one order of magnitude spread between the signal- and idler-photon energies. This is the range of extreme light, for which first applications have already been demonstrated.

In the following, the development of quantum-optical measurement concepts in extreme spectral ranges with a substantial spread of signal (detection) and idler (measurement) wavelengths is reviewed. Our aim is to present the state-of-the-art of the fields — namely the MIR and terahertz ranges on the low-frequency side of the visible spectrum as well as the UV and X-ray ranges on the high-frequency side — together with an overview of the theoretical descriptions. For a more detailed introduction to the field of nonlinear interferometry and path indistinguishability refer to refs. [21, 22] respectively. Also the perspectives of quantum imaging have been reviewed before.[23]

2. Generation of Correlated Photon Pairs

The key component of a nonlinear interferometer in extreme wavelength ranges is the source of correlated photons. A simple and common source is shown in Figure 1a. It consists of a nonlinear crystal and utilizes SPDC of laser light. SPDC was first theoretically predicted in the 1960s[24,25] and experimentally demonstrated shortly after.[26–29] A pump photon decays inside a nonlinear crystal into a correlated pair of lower energy photons due to interaction with vacuum fluctuations, which is illustrated in Figure 1b. Historically, the generated photons are called signal and idler. If the signal and idler photons share the same wavelength, the process is referred to as degenerate SPDC and if the two photons have different wavelengths, it is called non-degenerate SPDC. Taking both photons together, the term biphoton is used to account for the correlation. These biphotons have useful properties, as they are generated under conservation of energy as well as momentum. While the conservation of energy leads to a spectral correlation of the photon pair, the conservation of momentum leads to a correlation in emission directions. Additionally, such a pair is generated not randomly over the length of the crystal but almost at the same time.[30]

Therefore, measuring one of the photons generated due to SPDC allows for a direct gain in information of its partner. In other parametric processes like sum- (SFG) and difference-frequency generation (DFG), OPO, and OPA stimulated emission occurs (see Figure 1b). These methods have in common, that either a second light beam is irradiated (second pump or seed), or a cavity is build to profit from stimulated emission for amplification. The existence of an idler photon does not guarantee the existence of...
a signal photon, since the idler photon could have passed the crystal without interaction. Thus, the measurement of an idler photon does not allow a conclusion about a signal photon, as idler photons can originate from the seed laser or previous passes through the crystal due to a cavity. Also, the measurement of a signal photon does not lead to a gain of knowledge about an idler photon, because in stimulated processes after the conversion either two or no idler photons are present. Thus, there is no gain of information about a single partner photon, as in the case of photons generated by SPDC.

The correlation of photons generated by SPDC already forms the basis for the imaging technique called ghost imaging. In this imaging principle, the spatial and temporal correlation of the photons is primarily used. After generation, the correlated pairs are separated. The idler radiation illuminates an object and is afterward captured by a multi-pixel detector that only recognizes its time stamp. At the same time, the signal radiation is recorded by a multi-pixel (and therefore spatially resolving) detector which either records a time stamp itself or is only open for a short period of time triggered by the arrival of the idler. Thus, the time correlation is used to establish a relationship between the measured signal and idler photons. Since the photon pair is also spatially correlated, an image of the object can be determined from the signal photons that can be clearly assigned to an idler photon event. The advantage of this imaging method is that only a single-pixel detector is needed in the spectral range of the idler. These single-pixel detectors are usually much easier to implement. For a deeper insight into the field of ghost imaging, please refer to refs. [36–38].

In classical optics the development of almost every optical device starts with a simulation of its optical properties. As quantum sensing is a relatively young field, the corresponding simulations are not as developed. Correlated photon sources based on SPDC are an essential part of nonlinear interferometers. Therefore our overview of the simulation techniques starts with a deeper insight into the theoretical description and different concepts of SPDC-source simulation. The process of SPDC has been reviewed before, describing the semi-classical approach as well as the quantum-mechanical one. In contrast, our focus is on the evaluation of the different approaches for simulations and the reproduction of experimental results.

2.1. Simulation of SPDC Sources

The field of the theoretical aspects of SPDC sources is as diverse as the applications for entangled photons, since approaches for modeling sources can be vastly different, depending on the application. Here two different approaches are introduced and a number of references are presented that show the variety of models that can be obtained. These range from analytical descriptions to complex models that need to be solved numerically.

The theoretical descriptions are mostly independent of the specific wavelength regimes. However, there are some issues especially prominent at extreme wavelengths which are discussed later. The foundation of the theoretical work is applicable across a broad range of SPDC sources. Many of the references presented here do not explicitly deal with extreme wavelengths but their approaches can be used across regimes.

Two fundamentally distinct approaches have been developed for the simulation of SPDC sources. Both methods use different approximations and are therefore useful for different setups. In the following, the derivation of these methods to show the characteristics of the models is sketched. Further, a range of works where these methods have been applied to simulate a range of experimental setups is presented.

The first method is based on the second-order nonlinear part of the Hamiltonian for the electromagnetic field:

$$ H_{NL}(t) = \frac{1}{3} \int d\mathbf{r} c^{(2)}_{\mathbf{r}}(\mathbf{r}) D_{\mathbf{s}}(\mathbf{r}, t) D_{\mathbf{i}}(\mathbf{r}, t) D_{\mathbf{i}}^*(\mathbf{r}, t) $$

where $c^{(2)}_{\mathbf{r}}$ is the inverse susceptibility tensor and $D$ are the displacement fields. A formulation with the displacement fields instead of the electric fields is necessary to ensure consistency with Maxwell’s equations. The displacement fields for idler and signal $D_{\mathbf{i/s}}$ are then quantized by replacing them with field observables $\hat{D}_{\mathbf{i/s}}$. The pump beam is often bright enough to be treated classically. The undepleted pump approximation is often applied, since the probability of SPDC is small, such that it is reasonable to assume that the pump intensity does not decrease. The time evolution of the initial quantum state is approximated using first-order perturbation theory:

$$ |\Psi(t)\rangle \approx \left( 1 - \frac{i}{\hbar} \int_0^t dt' H_{NL}(t') \right) |\Psi(0)\rangle $$

where the initial state $|\Psi(0)\rangle$ is given as the vacuum state $|\text{vac}\rangle$ for signal and idler photons. Further adaptations and approximations can be made depending on the simulated setup. Typically, the integrals over time and space can be evaluated analytically. Given the biphoton state $|\Psi(t)\rangle$, the rate of biphotons produced in a certain mode can be calculated as the expectation value of the corresponding operator

$$ \Gamma(k_s, k_i) = \frac{1}{i} \langle \psi(t)| \hat{a}^\dagger(k_s) \hat{a}(k_i) \hat{a}(k_s) \hat{a}(k_i) |\psi(t)\rangle $$

Many other quantities can be obtained as well but may require integration over a number of modes. To calculate the single-photon count rate of the signal for example one needs to integrate over all idler modes leading to

$$ \Gamma(k_i) = \int dk_s \Gamma(k_s, k_i) $$

The specific model used varies largely across literature, mainly depending on the application. Models range from analytical expressions to high-dimensional integrals that need to be evaluated numerically. Depending on the requirements and the regime simpler models can be just as good as complex ones. However, a systematic analysis of these regimes and a comparison of model accuracy is not available. A comparison would aid researchers in picking an appropriate model based on a quantitative rationale. To point out the importance of simplified models, the example of calculating the distribution of signal photons which involves a six dimensional integral over the spatial domain and the idler momentum is employed. To calculate the integral, typically the integrand has to be evaluated $N^6$ times. Reducing the
complexity by simplifying the model can bring the computation time from the infeasible to the feasible region. A common example is the approximation of the square poling function of $f_p^2(r)$ with a sine function, yielding an analytical expression for the spatial integral in Equation (1).\cite{46} Thus, the three-dimensional integral does not have to be evaluated numerically, reducing the required integrand evaluations to $N^3$ and with it the computation time drastically.

One of the challenges this model provides is the balance of complexity and approximations. Increasing the complexity of models optimally leads to more accurate simulations but they also become more expensive to evaluate numerically as they involve higher-dimensional integrals. The photon momenta of the three-wave mixing process contain 9 degrees of freedom which are often reduced to make the problem more tangible. This can be done for example by limiting the wavelengths of the photons to a small range or a single value. The material parameters of many nonlinear crystals depend on wavelength, direction, and polarization, limiting the symmetries one could exploit to reduce the dimensionality of the problem. These, often nonlinear, relations also stand in the way of obtaining analytical solutions. As an example take a wave packet with a Gaussian wavelength distribution. After transition to the nonlinear medium its distribution will no longer be Gaussian due to the wavelength dependent refraction index.

Early descriptions used a plane wave approximation for the pump beam.\cite{30} The application to coherence properties of the SPDC photons showed that a reduced model could still predict various characteristics of the biphonon state.\cite{47} However, a transverse Gaussian profile represents experimental setups better and was shown to explain experimental data more accurately.\cite{48} Many recent simulations approximate the pump beam as collimated with a Gaussian transverse profile. This approximation allows for an analytical solution of the spatial integral in Equation (1) while reflecting important characteristics of many experiments. The pump focusing is then represented by the diameter of the collimated beam. With this model, focusing effects can be approximated by adapting the beam diameter.\cite{49} This approximation becomes less accurate as the focusing becomes stronger and crystals become longer. For strongly focused pump beams the variation in transverse momenta of the pump becomes significant and for long crystals the change in beam diameter over the crystal cannot be neglected.

The approximation of the characteristic sinc functions with other functions has been explored in many different ways.\cite{14,30,51} While a common choice is a Gaussian distribution, the choice for its width differs across literature. The Gaussian approximation also results in a simpler transfer function under the paraxial approximation and for monochromatic fields.\cite{41}

The pump walk-off is also subject to investigations.\cite{45,52} The azimuthal symmetry of the down-conversion spectrum is broken due to the asymmetry introduced by the pump beam. Ramírez-Alarcón et al.\cite{49} provide an analysis of this effect for different pump and crystal configurations. The numerical results for a number of degenerate SPDC spectra are shown in Figure 2a. The count rates were scaled to fit the experimental ones.

This model was also applied in the extreme light regime.\cite{54,55} Riewinger et al. demonstrate the application of this method to the creation of visible and terahertz photons. In this regime the influence of thermal photons plays a crucial role.\cite{56} The numerical results providing the absolute count rates for the visible light are shown in Figure 3c. Multiple quasi phase-matching (QPM) orders can be observed at the same time as well as photons at a wavelength smaller than the laser’s. These are created by an up-conversion process enabled by the existence of thermal photons.

The second approach to modeling SPDC sources is obtained along the lines of classical three-wave mixing but the seed laser is replaced by vacuum fluctuations. From Maxwell’s equations one can derive the wave equation

$$\nabla^2 E - \frac{1 + \chi^{(1)}}{c^2} \frac{\partial^2}{\partial t^2} E = \frac{1}{c^2} \frac{\partial^2}{\partial t^2} P_{NL}$$

(5)

where the slowly varying amplitude approximation was employed and $P_{NL}$ is the nonlinear part of the polarizability. Under the paraxial approximation the second-order derivatives can be neglected and one obtains a system of coupled equations for the field envelopes

$$i \frac{\partial}{\partial z} A_j + \frac{1}{2k_j} \nabla^2 A_j = \frac{\alpha_j}{k_j c^2} \chi^{(2)}(r) A_i A_i^* \exp(-i\Delta k z)$$

(6)

for $j = i, s$ and $k = s, i$. The spatial dependence of the field envelopes is omitted for better readability. The field envelopes $A_{j/k}$ are then quantized and replaced by their quantum operator counterpart\cite{58} or the expectation value of these operators.\cite{53} The pump field is treated classically in this approach as well and the undepleted pump approximation is employed. These equations provide an implicit expression for the quantities of interest such as first and second order correlation functions. A solution to these may be found using stochastic or Green’s function methods.\cite{58}

This coupled wave equation approach is valid across pump power regimes as higher-order effects are included in this model. An early adoption of this model shows the application to obtaining photon creation rates and correlation properties for multimode collection.\cite{59} The conversion rates to narrow-band single-spatial modes have been investigated as well.\cite{60} A comparison of the Green’s function method and Monte–Carlo simulations has shown that both options provide promising results for the simulation of experimental setups. The stochastic nature of the Monte–Carlo simulation leads to spatial statistical fluctuations similar to the ones observed in experimental images. The stochastic simulations also provide a handle to balance simulation time and accuracy.\cite{58} The simulation of irregularly structured crystals is a notable strength of this approach which can be used to show the conservation of orbital angular momentum in the SPDC process.\cite{53} With the first-order approach solving this type of problem requires the numerical integration over the spatial domain. Efficient simulations of complex poling structures enable the optimization of sources to obtain photon pairs with desired spectral and spatial properties. This has been shown for spectral\cite{61} and spatial shaping.\cite{62} Combining the two brings many more possibilities to improve SPDC sources for various applications.
Figure 2. Simulations of angular SPDC spectra. a) Angular spectra for a range of different crystal lengths and pump waists. Each row (e.g., subfigures (a–d)) shows the experimental (e.g., a) and simulation (e.g., b) results for one configuration with crystal length and pump waist decreasing from top to bottom. Reproduced with permission. [45] Copyright 2013, IOP Publishing. b) Angular distribution for degenerate SPDC process showing simulation and experimental results for different crystal angles, \( \theta \). Reproduced with permission. [53] Copyright 2020, John Wiley and Sons.

Figure 3. Simulations of frequency-angular spectra with idler photons in the terahertz frequency range. Experimental (solid curves) and simulated (dashed curves) spectral distribution of the collinear signal photons for a) forward- and b) backward-propagating idler photons. Reproduced with permission. [57] Copyright 2014, Springer Nature. c) Experimental and simulated frequency distribution of signal photons with an emission angle, \( \theta_s = 0.2^\circ \) considering multiple QPM orders. Reproduced under the terms of a CC-BY license. [55] Copyright 2022, The Authors.

The applicability of this model to real experiments was shown by simulations of SPDC sources and the subsequent optical setup. The first- and second-order correlation spectra of an SPDC experiment were reproduced using the stochastic approach and a split-step Fourier method. [53, 58] Results comparing the simulated with the experimental angular spectra are shown in Figure 2b. Multiple configurations were tested and a relative intensity was calculated. The results also demonstrate that using this method it is possible to simulate the transition regime between low and high-gain regime.

In the extreme light regime this method has been applied as well. Kitaeva et al. [57, 63] apply this method to the creation of photon pairs with the signal in the visible and the idler in the terahertz range. The effects of thermal photons and absorption in
the terahertz range were included. This allowed for the estimation of the spectrally resolved absolute count rates. The experimental and theoretical results for the collinear signal spectrum are shown in Figure 3a,b.

Both of the presented models are derived making different assumptions where the most notable are the first-order approximation for the first-order model and the paraxial approximation for the coupled wave equations. These different limitations make the methods applicable to different regimes. The first-order model is better suited for the low-gain regime, but is not limited to the paraxial range while the opposite is true for the wave equation model. The first-order model can be extended to include stimulated processes and higher-order photon interaction such that it is applicable to setups with higher pump powers. This would however lead to a large increase in complexity leading to potentially prohibitively high computational costs. Absorption effects in the crystal have only been modeled with the wave equation approach. These features make it clear that both models are useful in different regimes and provide a good extension to each other.

Simulating extreme light applications brings additional challenges. One difficulty is the large spread of wavelengths. Many applications have one extreme wavelength and one in the visible wavelength range. Taking the setup of Haase et al. for example, a small range of 5 nm in the visible corresponds to wavelengths from 100 \( \mu \)m to 1000 \( \mu \)m for the terahertz photons. This means the single-frequency approximation is often not applicable, especially for the larger wavelength. The wavelength spread also makes the wavelength dependency of material parameters more prominent. The same issue appears with the emission angles as a small angle in the smaller wavelength corresponds to a much larger angle in the larger wavelength. The transverse momentum conservation dictates a large emission angle for the lower-energy photons even for relatively small angles in the higher-energy photons. This can be approximated by \( \theta = \Theta \frac{\lambda_h}{\lambda_h} \), where the indices \( h \) and \( l \) stand for high and low energy, respectively, and the ratio of wavelengths will be large for this example. Other problems that occur especially in the extreme light regime are the influence of thermal light, absorption, and internal reflection in the nonlinear material. For periodically poled nonlinear materials the wavelengths of photons can be at the same order as the poling period which invalidates a common approximation. Due to the large wavelength spread sometimes multiple phase-matching orders can be observed at the same time. This provides additional challenges for modeling and increases computational costs.

The simulation of SPDC sources is a field as diverse as its applications. Significant progress has been made in recent decades. A multitude of methods from the simulation of absolute brightness, crystal temperature calibration to highly resolved spectra of single photons and coincidence measurements are at our disposal. With these tools SPDC sources for almost any application can be designed in detail before setting them up. Further work is necessary however to provide an efficient way to simulate highly resolved properties of SPDC sources. These simulations constitute the basis for modern applications of SPDC such as quantum spectroscopy and imaging. More systematic exploration is needed in comparing the different approaches and model variants. A better understanding of the appropriate regimes for the models would also help finding a model with a good trade off between accuracy and efficiency.

### 2.2. Sources of Correlated Photons with High-Frequency

The generation of correlated photon pairs based on SPDC with one partner in the X-ray or UV region is still subject of current investigations. Major challenges are the availability of suitable laser sources or suitable nonlinear materials. Therefore, in the following, hurdles of the individual fields in the generation of biphotons will be discussed and in addition, first realizations or demonstrations based on biphotons with pump sources in these spectral ranges will be given.

#### 2.2.1. X-Ray Radiation

The generation of correlated photon pairs in the X-ray range was first proposed by Freund and Levine, discussing the interaction of X-ray photons with atoms of a Bragg-layer structure. Such effects, based on interaction with vacuum fluctuations, had already been described for the optical region, and should also occur in the X-ray region. However, since the distance between the atomic lattice planes is comparable to the wavelengths of X-rays, the reciprocal lattice vectors play a decisive role in determining possible scattering angles of the biphotons to fulfill the phase-matching condition. This limits their angles to only small deviations from the typical Bragg-reflection criteria.

Additionally, the generation of correlated biphotons in this spectral region is hindered due the lack of easily accessible and coherent pump sources. To operate a down-conversion process in the X-ray range photon sources with even higher energies are required, since energy conservation must be satisfied. But especially for the case of photons with very high frequencies traditional laser gain media are not usable and alternative methods such as free-electron lasers, often require big facilities. In addition, there is only a very small selection of materials suitable for optical elements, as refractive indices are often close to 1 and the materials exhibit absorption. Even with special coatings or other optical techniques to overcome this limitation it is not yet possible to obtain optical elements with similar properties as in other spectral ranges. Therefore, X-ray lasers are often built without a laser resonator and only allow poor coherence properties. Since coherence is important in interferometric and correlation experiments the potential in spectral regions with high photon frequencies is limited.

Nevertheless, the generation of correlated photons with pump sources in the X-ray range has already been demonstrated. Although previous demonstrations of SPDC in the X-ray region are the subject of current discussion, an overview of the possible methods for generating and applying correlated photon pairs using PDC in this spectral range is given. In this chapter, we categorize photons by their energy rather than by their wavelength as this is the commonly used unit for X-ray photons.

The first demonstration of generating photon pairs in the X-ray was performed by Eisenberger and McCall shortly after the theoretical prediction. Using radiation at 17 keV, correlated photon pairs with single photon energies of 8.5 keV (wavelengths...
around 150 pm) were generated near the Bragg scattering criteria of a beryllium single crystal. Depending on the precise orientation of the beryllium crystal, the distance between the two detectors and the relative orientation of the detectors with respect to the generating crystal, up to one correlated biphoton per hour was measured.

But especially the generation of strongly degenerate biphotons, whose frequencies are in the X-ray and optical range, has been of great interest from the beginning. This could combine the strengths of both spectroscopic techniques of the optical domain and the high-resolution capabilities of X-rays. Additionally, detector limitations in the X-ray regime could be circumvented this way. A first demonstration with idler photons in the UV was achieved by Danino et al. This demonstrated the decay of 8 keV X-rays in a LiF single crystal into signal photons at 7.7 keV and corresponding idler photons in the extreme UV at 335 eV. In good agreement with theory, several hundred additional photons were observed at optimal phase matching than would have been expected from background signals only.

The first observation of wave mixing (sum-frequency generation) of X-rays and optical wavelength was achieved using a diamond crystal. In addition to X-rays at 8 keV the crystal was also illuminated with a Ti:sapphire laser at 1.55 eV (800 nm). After this door opener, several studies have been published investigating the properties of PDC in the near UV or even visible frequency range, even using standard laboratory X-ray sources. Today, these experiments achieve count rates with many thousands of down-converted photons per second, while generated idler wavelengths cover a broad range between 280–650 nm. The resulting idler wavelength is thereby mainly determined by the phase-matching condition, depending on the orientation of the crystal lattice with respect to the incident beam.

Up to now, mainly diamond single crystals have been applied to generate biphotons in the optical and the X-ray frequency range. Although conversion efficiencies of the order of 10−10 are reported, only a small fraction of the generated optical radiation is accessible due to internal reflection. But as recently demonstrated, other nonlinear crystal materials such as lithium niobate (LiNbO3) and gallium arsenide (GaAs) offer conversion efficiencies several orders of magnitude higher than any other material known to date. In particular, the lack of inversion symmetry appears to be the reason for this much stronger nonlinearity. In addition to the material, the polarization of the radiation also plays an important role in the efficiency of the down-conversion process.

However, at this point, it should be briefly stated why many of these experiments are currently under discussion. As a clear proof for PDC into optical regions, the characteristic scattering signature should be observable, resulting directly from the phase-matching condition. However, this signature is not reported in most experiments. Moreover, the determined conversion rates seem to be too high compared to other calculated and experimentally determined upper limits. Considering this, another possible explanation for the measured signal is elastic scattering at the apparatus itself.

Nevertheless, experiments in the X-ray regime mainly suffer from the low inelastic conversion cross section and the extremely narrow phase-matching conditions with small angular tolerances. In addition, many experiments use avalanche photodiode detectors (APD), that have low quantum efficiencies in this range. For this reason, the received count rate is often smaller than predicted by the theory based on the count rate equation.

### 2.2.2. X-Ray Ghost Imaging

Even though works on the generation of correlated photon pairs in the X-ray regime are still at the proof-of-principle stage and also subject of current debate, first experiments that exploit the correlation of these biphotons had already been realized. Thus, ghost imaging with degenerated biphotons in the hard or soft X-ray region was demonstrated. Similar to ghost imaging experiments in other frequency ranges, a mask is placed in the beam path of the idler photon (see Section 2). By measuring coincidences of a camera detector (signal) with a bucket detector (idler), the image of the mask can be reconstructed. The schematic of a common experimental setup for ghost imaging is shown in Figure 4a. Apart from this to the best of our knowledge, only quantum-optical measurement concepts based on the Mößbauer effect have been published in the X-ray range so far. However, since these experiments are not based on the generation of correlated photon pairs, they are not discussed in this review article.

The first realizations of ghost imaging in the X-ray range were not based on the PDC process but on the correlation of the light field itself similar to thermal ghost-imaging techniques. In one experiment, the natural fluctuations of thermal sources of electromagnetic radiation were used — as in Hanbury–Brown–Twiss interferometers. Therefore, a beam splitter generated two copies of the irradiated light field from a synchrotron source with energies of 20 keV. Using the intensity correlation of a bucket detector behind the object with the image captured by a camera, the shadow of the object had been recovered. In another realization, a monochromatic X-ray beam with 12.1 keV was used, exhibiting pseudothermal speckle-pattern fluctuation after scattering from a moving gold film. This pseudothermal beam hits the sample, which was moved in and out of the beam. To compute the image a Fourier transform was used. Recently, this ghost imaging scheme has also been extended into the regime of free-electron lasers.

Lately, ghost imaging based on biphotons generated by PDC has also been demonstrated. In Figure 4b a simplified schematic of such an experimental setup is sketched. With photon energies of 23 keV degenerate signal and idler biphotons in a diamond crystal at 11.5 keV were generated. The photons generated in this process have different emission angles such that the biphoton partners are easily separated into different paths. On their way to the bucket detector, the idler photons pass through a mask as in conventional experiments. However, in this experiment, the signal photons also passing a slit and are subsequently measured by another single-pixel detector. Since both the slit and the single-pixel detector are scanned through the signal beam path, this detector can be considered as a multi-pixel camera with pixel size equal to the used slit width. By coincidence measurements, the actual image of the mask was computed. For that calculation, only coincidence photon detections occurring at both detectors within 120 ns were considered, for which furthermore,
the sum of the idler and signal energy was equal to the pump energy within an energy window of 1 keV. A total of 20 to 80 coincidence events per pixel were recorded (depending on the size of the actual sample), while almost no coincidence events occurred in case the idler was blocked.

Although there is a very limited amount of works investigating external samples using nonlinear X-ray correlation and experiments are still at an early stage of fundamental research, findings such as the recent demonstration of ghost imaging based on biphoton correlation\textsuperscript{[92]} could open a pathway to medical imaging or nondestructive characterization which is particularly sensitive for high X-ray photon fluxes.

### 2.2.3. Ultraviolet Spectral Range

To the best of our knowledge, neither biphoton generation with one partner in the UV and the other in the visible, nor measurement principles based on biphotons with one partner in the UV spectral range have been demonstrated so far. For the generation of such a biphoton, the pump source must have a wavelength well below 300 nm due to the conservation of energy. Even though comparatively easily accessible laser systems exist here in contrast to the X-ray range, limitations are imposed by the optical properties of the nonlinear materials. Popular nonlinear materials for the generation of biphotons show strong absorption edges in this range. For potassium titanyl phosphate (KTP), lithium tantalate (LiTaO\textsubscript{3}), LiNbO\textsubscript{3}, or bismuth triborate (BiBO) these absorption edges are in the range of 260–350 nm.\textsuperscript{[103–108]} On the other hand, beta barium borate (BBO) shows its absorption edge only at 189 nm, but provides significantly lower conversion efficiency.\textsuperscript{[109,110]} In the future, new material compounds may offer the possibility of generating biphotons in the near-UV and visible range by providing absorption edges at even shorter wavelengths. For example new nonlinear material compositions such as CsSbF\textsubscript{6}SO\textsubscript{4} provide absorption edges already below 240 nm.\textsuperscript{[111]}

Another approach for generating correlated photons with one partner in the UV and the other in the visible is four-wave mixing. Decisive for this process is the third-order susceptibility of the nonlinear material offering lower conversion efficiencies compared to second order processes. However, it is advantageous for this process that no short-wave pump sources are required. Although four-wave mixing techniques have already been investigated,\textsuperscript{[112–117]} this process needs to be further developed to take advantage of UV–VIS photon pairs in future applications.

In addition, sensor technology in the UV range must also be further developed with regard to sensitivity and speed.\textsuperscript{[118]} Thus, in particular APDs show great potential for the future. Therefore, SPDC found its way into scientific investigation only for UV sources generating degenerate biphotons in the visible or NIR region so far. These can be used, for coincidence measurements between signal and idler radiation. Based on temporal and spectral correlation, the so-called Hong–Ou–Mandel effect can be observed in coincidence measurements between signal and idler radiation.\textsuperscript{[119]} If indistinguishable photons are superimposed at a beam splitter, the input photons leave the beam splitter in pairs. Thus, the coincidence rate measured at the two outputs decreases in contrast to the case where the photons are distinguishable. This is called the Hong–Ou–Mandel dip after the observers of the effect.

The generation of degenerated biphotons with pump lasers in the UV, usually BBO crystals\textsuperscript{[118,120–123]} or lithium iodate (LiIO\textsubscript{3}) crystals\textsuperscript{[124]} with thicknesses between 10 and 10 mm are used. As pump source serve frequency-doubled solid-state lasers\textsuperscript{[118,122–123]} or gas lasers.\textsuperscript{[120,124]} In first implementations, basic theoretical results of nonlinear interferometers were experimentally confirmed.\textsuperscript{[120]} An initial application of this principle was demonstrated in the measurement of the absorption spectrum of Nd\textsuperscript{111}-doped glass in the range of 800–900 nm\textsuperscript{[111]} and of Er\textsuperscript{3+}:YAG crystals at about 650 nm,\textsuperscript{[124]} as the absorption of the sample affects the rate and distribution of the coincidence events.

Another application is the characterization of UV laser pulses in the femtosecond range using SPDC autocorrelation.\textsuperscript{[118]} Due to the high absorption coefficients of the materials used, the usual technique for measuring the pulse duration by second-harmonic generation cannot be applied for UV lasers. Instead, the signal and idler photons generated by the UV pulse are used in conventional autocorrelation techniques to measure the pulse duration of the signal or idler radiation. Subsequently, the pulse duration of the original UV pulse can be determined. In addition, the coincidence measurements can also be used to calibrate detectors.\textsuperscript{[122]} In this case, the generated biphotons are efficiently detected with a photomultiplier and the efficiency of the detectors is analyzed by examining the coincidence rate.
2.3. Sources of Correlated Photons with Low Frequency

On the low-frequency side, sources of correlated photons are much easier to implement. Due to the lower energy of photons in the infrared or terahertz spectral range compared to the visible range, visible lasers can be used as pump sources for SPDC. These lasers are highly developed and offer high powers with low spectral widths and little noise. However, the range in which the generation of correlated photon pairs is possible is currently technically limited. Due to energy conservation, the signal photons are only slightly frequency-shifted with respect to pump if the idler photons earn low energies. As a consequence available filter and laser systems are not spectrally narrow enough to filter the pump radiation while transmitting signal radiation. Today, this limit is at the lower edge of the terahertz frequency range (200 GHz).\cite{106}

The nonlinear materials mainly used as sources of correlated photon pairs on the low-frequency side are magnesium doped (MgO:LiNbO₃) or undoped LiNbO₃ as well as KTP crystals. The great advantages of these materials are the high nonlinear coefficients and the comparatively easy handling, both in terms of production and periodical poling.\cite{112–117} Other materials such as gallium phosphide (GaP) or gallium arsenide (GaAs) also show sufficiently high nonlinear coefficients, but are much more difficult to handle, especially in terms of periodic structuring.\cite{128,129}

The fundamental constraint for the generation of correlated photon pairs via SPDC is the conservation of energy (see Equation (7)). The frequency sum of the generated photon pair must be equal to the frequency of the pump photon. This already determines all possible pairs. Which of these pairs are generated efficiently is determined by phase matching (see Equation (8)).

\[ \omega_p = \omega_s + \omega_i \]  
\[ k_p = k_s + k_i - k_\Lambda + \Delta k \]

Since periodically poled crystals are used in most of the following experiments in the MIR as well as the terahertz frequency range, this special case is already considered in Equation (8) and Figure 5. This so-called QPM technique offers the possibility to adjust the generated wavelengths by periodically structuring the nonlinear coefficient of the crystal.\cite{8,10–13} In a simplified picture, the phase difference of the waves involved in the nonlinear process is reset over and over again in a way that destructive interference does not occur. This can be achieved by varying the sign of the nonlinear susceptibility as a function of the crystal length. Therefore, a wave vector \( k_\Lambda \) representing the periodic structure of the grating has to be added in the phase-matching condition.

Since the representation of the generated signal is the same for the MIR and the terahertz spectral range, the focus here is on the latter, as additional effects occur in this range. In Figure 6d,e frequency-angular spectra of the generated visible signal radiation with corresponding idler photons in the terahertz frequency range are shown. Since this type of representation is not common, a short introduction is given. An exemplary experimental setup to obtain a frequency-angular spectrum of the signal is shown in Figure 6a. A nonlinear crystal is irradiated by a pump laser source (blue) and signal (green) as well as idler photons (red) are generated. The idler radiation is not considered.
after the nonlinear medium, since usually the camera does not respond in the extreme wavelength ranges of the idler. Otherwise, the idler photons can also be separated from the signal by using a dichroic mirrors, for example. The remaining pump radiation is separated so that only the remaining signal radiation is imaged onto the camera. To study the wavelength as well as the angular distribution the method of crossed dispersion is used. To acquire sharper images, a slit is placed in the beam path to limit the transmission of rays with large horizontal wave vectors (see Figure 6b). Afterward, the signal is imaged by a lens through a spectrograph, usually a grating or a prism, onto the camera. Due to the spectrograph, the spectral components are distributed on one axis of the camera, while the angular distribution is directly imaged by the lens onto the other axis. This leads to the typical tails sketched in Figure 6c.

Figure 6d shows the frequency-angular spectrum of an unpoled LiNbO₃ crystal pumped at 514.5 nm. In Figure 6e a smaller section of the frequency-angular spectrum of a periodically poled LiNbO₃ crystal pumped at the same wavelength is shown. Only the horizontal V-shaped curves are due to the nonlinear conversion of photons. The prominent vertical line at an idler wave number equal to 0 cm⁻¹ is the remaining pump radiation due to the imperfect filtering. Additional vertical lines observable in Figure 6d correspond to radiation of polariton branches.

In contrast to other parametric conversions like DFG, injection seeded OPA or OPO, which have an external restriction of the generated wave vectors (e.g., by an external cavity), only the wave vector of the pump photons is predefined in case of SPDC experiments. As there are also no seed photons determining the wave vector of the signal or idler photons for noncollinear phase-matching the emission angles vary with spectral distance to the pump. In case of the periodically poled crystal in Figure 6e a special feature of the conversion to the terahertz spectral range can be observed. The wave vector defined by the grating of the periodically poling (see Figure 5) only provides an orientation and not a direction.⁹¹²,¹³⁻⁴ As a result, the vector in the phase-matching scheme can also take a direction opposite to the pump. In the case of terahertz radiation this leads to possible phase matching with so-called backward terahertz photons, emitted in the opposite direction with respect to the pump photons. As can be seen in Figure 5, the wave vector of the terahertz radiation is shorter in the backward-facing case than in the forward. Thus, the tails closest to the pump belong to the backward-direction and are not due to higher orders of phase matching.

A major difference to the experiments in the infrared is also the observation of up-converted signal radiation. Referring to Raman spectroscopy, the up-converted signal radiation (negative wave numbers in Figure 6d,e) is often called anti-Stokes radiation, while the down-converted signal (positive wave numbers in Figure 6d,e) is called Stokes radiation. In addition to SPDC, parametric conversion of thermal terahertz photons also contributes to the generation of signal photons. In contrast to higher-energy photons, there is a sufficient amount of thermal photons already at room temperature. As can be seen in Figure 6d,e, this leads to a higher number of photons in the Stokes part of the frequency-angular spectrum, since both SPDC and parametric conversion of thermal terahertz radiation contribute here, whereas in the case of up-conversion only conversion of thermal radiation contributes. Novikova et al., were able to demonstrate this experimentally, by cooling the crystal down to 4.2 K.¹³⁵ Then, thermal radiation no longer contributes to the conversion and only the process of SPDC can be observed.

2.3.1. Generation of Broad SPDC Spectra

Especially in context of spectroscopy and optical coherence tomography (OCT), it is important that the used sources emit very broadband radiation in order to be able to investigate large spectral ranges. For this purpose, the phase-matching conditions can be set either via the nonlinear material or the input parameters.

A simple way to broaden the spectrum by touching the nonlinear medium is to reduce the interaction length. This allows for much greater phase mismatch of the beams involved. Since the number of possible phase-matching conditions depends on the phase mismatch, broad spectra can be obtained this way (see Equation (8)). Okoth et al., have taken this to the extreme by using a layer of lithium niobate only 6 µm thick, resulting in a spectral width of the generated photon pairs of about 500 nm.¹³⁶ Additional work has also been done using films of gallium phosphide⁹¹⁷ or optical metasurfaces⁹¹⁸ allowing higher generation rates. However, the radiation generated by this method is not easily accessible as it is emitted under large angles.
In addition, methods have been developed that allow broader spectra of the generated radiation by a certain type of structuring of the nonlinear material, such as the use of crystals with aperiodical poling.\cite{61,139,140} For these, the width of the poling structure varies as a function of the crystal length. Since different phase-matching conditions exist in each region due to the different structuring, there is a broadening of the emitted spectrum. For example, this allowed bandwidths of up to 800 nm to be achieved in stoichiometric lithium tantalate.\cite{140}

However, for both methods mentioned, the broadening of the spectrum is accompanied by a reduction in the number of photon pairs generated due to a shortened interaction length. However, using ultrathin films allows to choose materials with very high nonlinear coefficients, since the photons hardly experience any scattering or absorption and the necessary coherence length is low.

Another approach to broaden the generated spectrum by the crystal properties is the generation of biphotons via non-collinear phase matching.\cite{141} This allows the effective group velocity of the signal to be matched with respect to the pump wavelength along the optical axis. Using this approach, degenerate SPDC spectra with a bandwidth of up to 200 nm were generated. Recently, also the combination of non-collinear phase matching and aperiodic structuring was demonstrated.\cite{142} The obtained spectrum contained idler wavelengths between 3.5 and 5.6 μm.

The spectrum of the SPDC can also be broadened via the pump radiation as input parameter. For example, using a spectrally broad pump beam leads to broader idler spectra as long as the phase-matching conditions of the material are fulfilled.\cite{143} Another technique is to use a tightly focused pump beam. Due to the strong focusing, many different pump wave vector orientations are presented and thus lead to a larger number of possible phase-matching configurations.\cite{144} However, in these experiments, emission of the generated radiation occurs also under large angles and, especially in case of the focused pump, the achieved spatial correlation is poor.

In 2019, Vanselow et al., proposed and demonstrated a method which can be used in particular in the MIR for broadening the generated biphoton spectrum by the choice of the pump wavelength. This method is based on the typical characteristics of the phase-matching conditions of nonlinear crystals, such as KTP and MgO:LiNbO\textsubscript{3}.\cite{145} These materials show a comparatively large slope of the refractive index in regions near an absorption band as in the UV or the far IR, as shown in Figure 7. Between these absorption windows, the refractive index is comparatively flat and the associated group velocity shows an extremum. Therefore, a broad range of MIR wavelengths is usually accompanied by wavelengths in the visible range with matching group indices. This is crucial for fulfilling the phase-matching conditions, since the group velocities of the involved beams must be the same. Using this method, in a periodically poled KTP crystal (ppKTP), a bandwidth of 1200 nm with idler radiation between 3250 and 4450 nm was experimentally demonstrated.\cite{145} In another realization for periodically poled LiNbO\textsubscript{3}, a 900 nm bandwidth of idler photons between 3100 and 4000 nm was generated.\cite{146,147}

In case of KTP the generated bandwidth of idler photons is limited by absorption of the crystal material. Overall, both LiNbO\textsubscript{3} and KTP exhibit similar transparency regions of 0.4–5.0 μm in the case of LiNbO\textsubscript{3}\cite{103} and 0.4–4.3 μm in the case of KTP.\cite{105}

Therefore, other materials with larger transparency regions such as silver thiogallate (AgGa\textsubscript{2}) are gaining more attention and may find application in the near future.\cite{148,149} Even though this method is very well suited for nonlinear spectrometers in the IR range, it cannot simply be transferred to other wavelength ranges, such as the terahertz frequency range. Since these wavelengths already lie within the absorption band of the considered crystals, a broadband matching of the group index with radiation in the visible range cannot be achieved this way.

It should also be mentioned here that recently the shape of the emitted SPDC spectrum is of increasing interest. Crystal superlattices with several alternating periodically poled and unpoled regions are used. For example, in such a LiNbO\textsubscript{3} crystal, a comb-like emission spectrum of signal and idler photons with large spectral bandwidths were observed.\cite{150}

### 3. Nonlinear Interferometry with Extreme Light

After the discovery of the SPDC process and the correlation properties of the generated photons, it took until the 1980s before these new sources were applied to optical coherence experiments.\cite{119,151} In this context, Zou, Wang, and Mandel investigated two of such nonlinear processes taking place one after another and discovered that the overlapped signal radiation of two SPDC sources pumped by a single coherent laser experiences interference under certain circumstances.\cite{18,19} Signal interference only occurred if the idler paths of both crystals were completely superimposed. This was achieved by aligning the idler radiation of the first crystal such that it passes through the second crystal. Since the idler photons from the first crystal did not cause additional emission in the second crystal but — in a semi-classical picture — induce a coherence between the two signals, this effect was called “induced coherence without induced emission.”\cite{19} Nonlinear interferometers based on “induced coherence” are nowadays often referred to as Mandel interferometers. Since in this work only nonlinear interferometers based on this principle are discussed, the term nonlinear interferometer is used synonymously.

A common quantum-mechanical explanation of the observed interference is based on the access to which-path information of the signal photons. This information is lost by overlapping the idler beams as it is not possible to determine whether a specific signal photon was generated in the first or second crystal by...
measuring the corresponding idler photon after the second one. The indistinguishability of the paths leads to interference of the signal beams as the possibilities of a signal photon in both paths interfere. Therefore obtaining information about the origin of the signal photons results in the loss of interference as the possibility for a signal photon in the second path is removed.\(^{[13]}\) To demonstrate this, a neutral density filter was inserted into the idler beam path between the crystals, inhibiting the interference of the signal radiation (see Figure 8a).\(^{[19]}\) In addition to the visibility also the phase of the observable interferogram is affected by the sample in “induced-coherence” experiments. However, changes in the interference not exclusively depend on the transmission or phase changes introduced in the idler path, but also on changes made to each of the other beams involved (pump and/or signal).\(^{[120]}\) But combined with the possibility to adjust the generation of the correlated photons, an elegant way of measuring phase-sensitive information in one spectral range and detecting information in a different one emerged. However, at this early stage of development nonlinear interferometry could not leap from a physically interesting phenomenon to application.

In order to exploit the full potential of this new method, further developments in two areas were essential. On the one hand, the detectors in one particular wavelength range (signal) had to be much further developed compared to those in other spectral ranges (idler). On the other hand, nonlinear sources with a high conversion efficiency had to be found. With regard to detectors, this development has taken place primarily in the visible spectral range. Due to the high consumer demand, detectors based on complementary metal-oxide semiconductor (CMOS) and charge-coupled device (CCD) technologies nowadays achieve high photon yields and are less expensive. Nonlinear sources, are still of great interest in optics as they allow parametric conversion to many spectral ranges with a large number of available crystallographic systems.

After the first demonstrations, the next major milestone of the measurement principle based on “induced coherence” took almost 20 more years. In 2014, Lemos et al. revived and extended the idea of indirect measurement to imaging and called it “imaging with undetected photons.”\(^{[152]}\) In this work, the basic principle of the effect already observed in refs. \(^{[18, 19]}\) was supplemented with spatially resolved detection. Since this experiment has been inspirational for a large variety of others, we elaborate the technical details in the following. By pumping two ppKTP crystals coherently at 532-nm pairs of signal (810 nm) and idler (1550 nm) photons were generated in each crystal. Similar to refs. \(^{[18, 19]}\), a stable interference between signal photons was observed after superposition by a beamsplitter when the idler radiation of the first crystal is guided through the second. But in contrast to previous experiments, the spatial correlation of the photon pairs was exploited as well. As demonstration, an object (cat outline) was placed in the idler path introducing a phase shift of \(\pi\) for idler photons passing through the cat outline. As can be seen in Figure 8b the object was clearly visible in the interference of the signal. Due to the spatial correlation of the generated photon pairs, manipulation of idler photons emitted at a given angle only affects corresponding signal photons.

This was the first time images were taken based on nonlinear interferometry detecting only the signal radiation, which never interacted with the object. In addition, the used object was opaque to the signal and the deployed camera was blind to the idler radiation. Overall, this demonstrated the great potential of nonlinear interference experiments and provided the impetus for obtaining spatially resolved, phase-sensitive images of objects in spectral regions which are difficult to access. In follow-up publications, the influence of misalignment\(^{[153]}\) and pump beam waist\(^{[154, 155]}\) on the spatial correlation of the photon pairs was analyzed, as these properties are relevant for obtaining sharp and high-contrast images.
Nonlinear interferometers using signal and idler photons in the visible and near-infrared, respectively, have been demonstrated to date for a variety of applications such as imaging, ellipsometry, and optical coherence tomography. In the following, the focus will be on nonlinear interferometers with a more extreme wavelength spread between signal and idler photons. In addition, the scope of this work is limited to low-gain nonlinear interferometers, for which a linear dependence of the signal visibility on the transmission of the idler photons is obtained. In contrast, high-gain variants provide a nonlinear dependence due to induced emission.

Since the geometry of nonlinear interferometers is as diverse as for conventional interferometers, a short overview of different geometries and their advantages and disadvantages will be given before the theoretical description. In Figure 9 various geometries of nonlinear interferometers are illustrated. If possible, the position of the object under investigation is indicated by a dashed line. An inserted object has two main effects on the observable signal interference: the visibility of the observed interference can decrease due to absorption and scattering and a phase shift can be induced due to a change of the optical path length. After superposition of the generated signal beams, detection can be performed following the dotted green line.

The most straightforward way to build a nonlinear interferometer is in compact Mach–Zehnder geometry (see Figure 9a), where two nonlinear crystals are placed around the object under investigation. In both crystals signal and idler photons are generated able to interfere with each other. The great advantage of this geometry is given by its uncomplicated alignment. Only the path between the two crystals must not be too long, so that the principle of indistinguishability for signal and idler photons emitted under a given angles still applies. However, this geometry is subjected to several severe limitations. First of all, the samples must be opaque to signal and pump radiation. Additionally, optical properties of the sample and phase-matching conditions of the biphoton generation must be known in advance. Moreover, intense pump radiation may damage the sample and scattering of pump as well as signal radiation may occur. Therefore, usually only gas samples are investigated with this interferometer geometry.

All disadvantages of the compact Mach–Zehnder geometry can be eliminated if only the idler radiation interacts with the object. In the Mach–Zehnder geometry, shown in Figure 9b, the idler photons are separated from the signal photons and are directed into the second nonlinear crystal. This allows for a large variety of samples even when no prior information in spectral regions of the pump and signal radiation is available. However, the complex alignment of the experiment is disadvantageous. In addition to plenty of optical elements, the path length difference has to be adjusted within the coherence length of the generated biphoton pairs. The path length difference of the interferometer arms must be matched within a few hundred micrometers. By using additional variable path lengths, this problem can be reduced.

Figure 9. Various nonlinear interferometer geometries. The object under test (dashed line) is placed in the idler path. a) Compact Mach–Zehnder geometry. b) Mach–Zehnder geometry. c) Young’s geometry. d) Michelson geometry.
Young’s geometry is another easy-to-implement geometry for nonlinear interferometers and is shown in Figure 9c. Following the famous double-slit experiment of Young, a double-slit mask is placed directly at the front facet of the crystal.[120] This way the crystal is pumped by two spatially separated beams and biphotons are generated in different regions within the crystal. For this geometry neither path lengths nor the superposition of beams need to be adjusted. However, it only allows studying the non-linear material itself, since the interference is modulated by the optical properties of the crystal.

Nowadays, nonlinear interferometers are typically implemented in Michelson geometry (see Figure 9d). After a first passage through the nonlinear crystal, all radiations involved get reflected and re-enter the same crystal. Similar to the Mach–Zehnder design, idler photons are separated from the signal and pump, and exclusively illuminate the object. Interference then takes place between signal photons generated by the first and the second pass. Intensity variation can be observed as a function of photon paths will be discussed. The focus is on interferometers in the low-gain regime as these are based on the quantum nature of photon entanglement.[13] High-gain variants are also available and work by means of induced emission.[165] When an object is inserted into the idler arm, the paths become more distinguishable and the interference in the signal changes. The distinguishability can be influenced by large phase shifts or by a reduced transmission caused by the object in the idler arm resulting in a decrease of visibility for the interference signal.[18,19] The theory was developed in parallel to the experimental applications such that the history is quite similar as laid out above. The theory was extended from the collinear region to a range of emission angles such that the formation of images can be described.[64] At each point of the detector the measured signal depends on the transmissivity and optical path length at a corresponding spot on the imaged object. Taking the Mach–Zehnder setup shown in Figure 9b, the photon count rate for a point \( \rho_{k_i} \) in the detector plane can be expressed as:

\[
R(\rho_{k_i}) \approx \Gamma_1(k_i; k_s) + \Gamma_2(k_i; k_s) + 2 \sqrt{\Gamma_1(k_i; k_s) \Gamma_2(k_i; k_s)} T(\rho_{k_i}) \cos(\Delta \phi(\rho_{k_i}))
\]

where \( \Gamma_1(k_i; k_s) \) are the photon count rates for the crystals NL1 and NL2 respectively, \( T(\rho_{k_i}) \) and \( \Delta \phi(\rho_{k_i}) \) are the transmissivity and phase shift of the imaged object at a point \( \rho_{k_i} \) that depends on the idler momentum \( k_i \). Here a perfect imaging system and perfect correlation between signal and idler momenta is assumed to simplify the equation. The third term is an interference term, where the strength of the interference depends on the material properties of the object. Due to this dependence they can be inferred from the detector image. The description of a setup with a Michelson configuration works along the same lines. A notable property of this kind of interferometer is that the magnification depends not only on the lenses but on the ratio of wavelengths as well. The theoretical description given here is based on the first order model of SPDC sources. A plane-wave decomposition of signal and idler allows isolating the modes that contribute to a single point in the detector plane.

Assuming perfect correlation between signal and idler allows deriving analytical results which qualitatively describe the effects observed in an interferometer.[64] For a quantitative description the influence of imperfect correlations between signal and idler needs to be considered.[48] Imperfect correlation means that for signal mode \( k_i \) the idler mode is not fixed, but instead a range of idler modes can be created following a distribution \( p(k_i; k_s) \). The entangled idler momenta from this distribution are propagated to different points on the object such that even in the absence of limiting classical effects there is no point to point correlation of a signal mode on the detector plane and a point on the imaged object.[164] The signal on a detector point instead represents a convolution of the area covered by these idler modes.[16] This property limits the resolution achievable with this method. The resolution limit is determined by the wavelength of the light that illuminates the object. The absolute size of the image of a point object is determined by the detected wavelength.[153] In some cases the distribution of idler momenta can be approximated by taking the transverse correlation function as a Gaussian distribution and considering a single wavelength for each idler and signal. This allows analytically calculating the limits of some properties of an interferometer.[155,164]

Effects such as misalignment of the optical system diminish the quality of the interferometer output. One cause is the variation of phase shifts within the idler distribution \( p(k_i; k_s) \). The principle of this effect has been investigated using an angle dependent phase which can emulate a longitudinally shifted lens.[164] The resulting interference pattern is shown in Figure 10b,c. Real setups have the additional issue that the alignment of the signal and idler rays is imperfect and therefore the path becomes partially distinguishable leading to a decreased visibility. The influence of both effects on an interferometer setup can be analyzed with a numerical model for a Michelson-geometry interferometer.[18] Figure 10a shows the result of such an analysis that also includes imperfect correlations.

Most published experimental works so far used a far field interferometer configuration, in which imaging is based on the correlation of transverse biphoton momenta. However it is also possible to use a near field configuration where the image formation is enabled by the position-correlation between signal and idler photons. An experimental realization has been published recently, showing the potential of this configuration for wide field microscopy.[165] The theoretical description has been developed.
recently as well, showing that the material parameters can be inferred from the detector image in a manner similar to the far field setup. However theoretical and numerical analysis of the resolution limit has shown fundamental differences. The resolution limit for the near field configuration depends on the sum of the wavelengths of signal and idler instead of their ratio. Further research on the imaging properties depending on the idler wavelength has been done and shown that the resolution becomes worse as the idler wavelength increases as the number of spatial modes available for imaging is reduced.

The theoretical description of quantum spectroscopy and similar schemes is based on the same principle as quantum imaging which is the indistinguishability of paths. For this application instead of the transverse momentum correlation the wavelength correlations is used. Further, spectroscopy requires broadband sources such that different approximations are required to derive the theory of quantum spectroscopy. Let us imagine a Michelson setup as shown in Figure 9d and add a translation stage to the idler mirror on the bottom left. The count rate for the collinear direction in dependence of the stage shift $\Delta l$ and the signal frequency $\omega_s$ can then be described as

$$R(\Delta l; \omega_s) \approx \int d\omega_i \left[ 2\Gamma(\omega_s, \omega_i) + 2\Gamma(\omega_s, \omega_i) T(\omega_i) \cos(\Delta \phi(\omega_i) + \frac{\omega_i}{c} \Delta l) \right]$$

(10)

where $\Gamma(\omega_s, \omega_i)$ is the photon count rate for the crystal, $T(\omega_i)$ and $\Delta \phi(\omega_i)$ are the transmissivity and phase shift of the object for the idler frequency $\omega_i$. The term $\omega_i \Delta l/c$ represents the phase shift caused by the shift of the translation stage. For the Michelson setup one needs to consider that the idler beam passes through the object twice and reflect this in the inferred properties. To derive this simplified equation again a perfect imaging system is assumed and the transverse momentum of the photons is neglected. It can be seen again that the properties of the object can be inferred from the interference pattern. The count rates typically depend on a squared sinc term. The integral over the interference term thus gives an interference pattern with a triangular envelope. This property has also been explained by the auto-correlation function of the SPDC photons which yield a similar triangular envelope. Using numerical simulations the results of complex experimental setups can be explained. This has been demonstrated for layer-thickness measurements with the idler in the terahertz regime. The simulations included the full spectral range of the SPDC photons and further the limiting apertures for the idler to reproduce the collinear interference pattern of the signal. Quantum time-domain OCT can be described with a very similar approach.

While qualitative theoretical explanations of the observed interferometer properties are available, the field of quantum based interferometer simulation is sparsely researched. The challenges for simulating this type of interferometers are the same as for the SPDC sources as they are an essential part of the interferometer. The computational requirements are even larger as additionally the 6D biphoton states need to be propagated through the imaging system and to obtain the interference pattern the degree of distinguishability between the two paths needs to be determined. The availability of interferometer simulations would be a useful tool to explore the limits of quantum based interferometers and to set a goal for experimentalists. Further, limiting components could be identified and replaced optimizing setups for performance. The robustness to confounding factors could be examined and improved as well which is an important step in designing devices fit for industrial applications.

### 3.2. Spectroscopy

Especially in case of the experimental realization of spectroscopy the concept of nonlinear interferometry has made a tremendous progress in recent years. Particularly for idler photons in the low-frequency extreme spectral regions as the MIR or the terahertz frequency range. This fast development is based on the expertise already gained in the field of nonlinear optics for these spectral ranges, whereas for high-frequency ranges, such as the UV or the
X-ray range, this fundamental knowledge has not yet been accumulated and needs to be built first. Therefore, only the progress and achievements of the concept with idler photons in the MIR or the terahertz spectral region are given below.

3.2.1. Mid-Infrared Frequency Range

The MIR is often specified to wavelengths from 3 to 50 μm. Here, many substances own characteristic spectral features, so-called fingerprints, like fundamental absorption bands of gases or the vibrational spectra of molecules. These features make MIR spectroscopy one of the most important techniques for the analysis of substances with wide application in industry and science. Therefore, typical measurement methods like Fourier transform infrared spectroscopy (FTIR) using MIR detectors are continuously developed. However, compared to optical detectors, MIR detectors usually show lower detection efficiencies, higher dark count rates, and additionally often require cooling. For this reason, many concepts based on nonlinear conversion have been developed in recent years shifting detection to the NIR or even the visible spectral range allowing the use of silicon-based detectors or InGaAs detectors.

In the following, nonlinear interferometry approaches based on “induced coherence” are discussed. To ensure the comparability of the individual works, the generated bandwidths in the idler range and the achieved spectral resolutions are given in the unit cm⁻¹ typical for spectroscopic investigations.

In most demonstrations of nonlinear interferometry focusing on spectroscopy in the MIR, the nonlinear material used is MgO:LiNbO₃ pumped at a central wavelength of 532 nm. Phase matching is achieved either via critical phase matching dependent on the angle or via QPM. In the case of critical phase matching the applied crystals have lengths up to 1 mm, allowing spectral bandwidths between 50 and 150 cm⁻¹ to be realized for idler photons at 4.2 μm. Slightly varying the angle changes phase matching and measurements over a spectral range exceeding 500 cm⁻¹ are achieved. Using QPM, the emitted bandwidth can be further increased achieving bandwidths of more than 100 cm⁻¹ using a 10 mm crystal length. Varying poling period and crystal temperature, almost the entire range between 3.2–3.9 μm (560 cm⁻¹) was covered.

In order to achieve very broad spectra of idler photons in the MIR without varying these parameters while performing the experiment, the method proposed in ref. [145] (Section 2.3.1) is used. For periodically poled LiNbO₃ crystals, this requires a pump wavelength of 785 nm, allowing a bandwidth of idler radiation of 725 cm⁻¹ to be achieved at 3.6 μm. However, the corresponding signal photons with wavelengths at 1 μm are no longer in the visible range. There silicon-based detectors generally show low quantum efficiencies. The experiments with KTP are based exclusively on broadband phase matching and are performed at a pump wavelength of 660 nm. Here, an even larger spectral bandwidth of 850 cm⁻¹ of the generated idler photons at 3.8 μm was realized with corresponding signal photons at 800 nm.

Considering instrumentation, a wide range of stocked optics can be used to filter the pump radiation with respect to the signal radiation. It can either be achieved by the application of phase matching with crossed polarization or by dichroic optics as mirrors, notch filters, or long-pass filters. For many geometries of nonlinear interferometers it is also necessary to separate idler and signal radiation. Due to the use of idler photons in the MIR and signal photons in the visible or NIR, expensive custom made optics are often required, especially since losses in this region of the interferometer are at the expense of the observable interference visibility. Typical achieved visibilities range from 14% to 30% and do not reach the demonstrated visibility values with idler photons in the NIR.

The detection of the signal interference can be applied by the method of crossed dispersion (see Figure 6a), resulting in an observable interference for the individual wavelengths as a function of the emission angle. However, due to the input slit, a large part of the interfering signal is eliminated and additional losses at optical elements (grating/mirror) of the spectrograph occur. Therefore, it can be advantageous to image the generated signal radiation directly onto a camera. Omitting the spectrograph results in a signal spectrum purely depending on the emission angle, as can be seen in Figure 6b. In this case, the interference pattern is displayed in radial direction for different emission angles and can be modulated, for example, in Michelson geometry by changing the path length difference. It is also possible to measure a spectrum purely depending on the wavelength by detecting the signal radiation directly by a fiber-coupled spectrometer (pixel line). In this case, the observed interference results as a function of the wavelength. In addition to multi-pixel detectors, the usage of single-pixel detectors without spatial resolution can be useful. The radiation can either be focused directly onto a photodiode or passing a monochromator before. Since spatial interference is no longer observable in these cases, the modification of interferometer parameters is mandatory for observing interference.

The first realization of spectroscopy based on nonlinear interferometry with idler photons in the MIR was performed in 2016 by Kalashnikov et al. At the same time, this was the first realization of spectroscopy based on this concept. In compact Mach–Zehnder geometry, measurements of the absorption coefficient and refractive index of CO₂ gas in a chamber as a function of pressure was demonstrated (see Figure 11a,b). The study focused on a strong absorption at a wavelength of 4.27 μm. The recorded frequency-angular spectrum was modulated by the emission angle at a fixed wavelength due to nonlinear interference. By fitting the observed interference signal with and without the gas, the parameters of the gas were determined over a spectral width of 500 cm⁻¹, matching roughly the calculation. Due to the used spectrometer, a resolution of 20 cm⁻¹ was achieved.

Omitting the spectrograph, a much higher signal-to-noise ratio of the observed interference was achieved by averaging the angular spectrum at fixed radii. However, with similar accuracy of the determined parameters, the obtained resolution is four times lower than the resolution limited by the natural linewidth of the generated biphotos in this case.

Recently, the method demonstrated in ref. [176] was modified using a crystal superlattice. Therefore, additional nonlinear crystals (total number of five crystals) were placed in the gas...
and phase shift of CO$_2$ agree well with theoretical simulations. Performed measurements of the absorption coefficient of various samples. The determined parameters matched well with literature or measurements with a commercial system.

An estimation of refractive indices and transmission at various wavelengths allowed for the calculation of the reflection and the absorption coefficient of various samples. The determined parameters matched well with literature or measurements with a commercial system.

A similar approach was realized for idler photons at 3.8 $\mu$m covering a bandwidth of 850 cm$^{-1}$. Using a fiber-coupled line-pixel spectrometer a spectral resolution of 1.5 cm$^{-1}$ was achieved. Performed measurements of the absorption coefficient and phase shift of CO$_2$ agree well with theoretical simulations and even show the fine structure.

In addition, Kaufmann et al. demonstrated an approach focusing on speed, without the need for varying path lengths during the measurement. For that, the interferometer is adjusted far from balanced arm length producing high-frequency interference over the observable spectral bandwidth. Using a Hilbert transform, the amplitude as well as the instantaneous phase over the whole spectral width was calculated. By referencing to a measurement without a sample, the transmission of various polymers was determined with measurement times of 1 s for each spectrum, reaching the acquisition rates of classical systems. In comparison, other demonstrations require measurement times in the range of several minutes to record similar bandwidths, however, higher resolutions are obtained. The resolution of this method is mainly determined by the observable carrier frequency of the interference. Ideally, this oscillation is of higher frequency than the characteristic size of resolvable features. Considering the smallest structures observed, a resolution of about 12 cm$^{-1}$ is given for this approach.

In analogy to classical Fourier transform spectroscopy, Lindner et al. demonstrated measurements without spectrally selective detection. In this case, the spectral information is obtained exclusively by a Fourier transform of the interferograms recorded via variation of the path length difference. Since the spectral information is obtained by a Fourier transform from the spatial domain, the spectral resolution is determined solely by the maximum feasible shift. The available shift of 800 $\mu$m results in a resolution of 6 cm$^{-1}$. By using different phase-matching configurations, the transmission of a polypropylene film (absorption at about 3.4 $\mu$m) was measured almost over the entire bandwidth of 560 cm$^{-1}$ around (3.2–3.9 $\mu$m).

In a subsequent improvement, a high bandwidth of 725 cm$^{-1}$ (3.1–4.0 $\mu$m) was realized by adjusting the pump wavelength in a single phase-matching configuration. Due to the associated shift of the signal photons to wavelengths around 1 $\mu$m, a silicon APD is used for detection. As before, the recorded interference is transferred via a Fourier transform. By implementing a longer variation of the arm length of 20 mm, a spectral resolution of 0.56 cm$^{-1}$ was achieved. To characterize the system, the transmission spectrum of methane was measured in the range of 3.3 $\mu$m, resolving rotational lines. By apodization an accuracy of the measured transmittance value of 1% was achieved (see Figure 11c). Recently, also the dispersive properties of the gas sample have been retrieved.

### 3.2.2. Terahertz Frequency Range

In the previous section experiments and interesting applications of quantum spectroscopy in the MIR-frequency range have been discussed. Although this frequency range already benefits from silicon-based detectors due to their low noise threshold, others can profit even more. In particular, the terahertz frequency range can gain from this new measurement technique. Although commonly used components have undergone tremendous development in recent decades, detection in this frequency range is still very challenging. Due to the low photon energy of about 4 meV, the detectors are technically complex and often require strong improvements.
cooling. Additionally, most commercial systems use single-pixel detectors which only allow for slow raster scanning and are rather expensive due to the laser systems which they are based on.

However, the application of nonlinear interferometry in this spectral range is also subject to technical challenges. Due to the low energy of the generated terahertz radiation, there is only a little spectral separation between signal and pump radiation (e.g., 1 THz difference at 532 nm corresponds to a shift of about 1 nm). Therefore, extremely narrow bandwidth lasers and filters have to be used. Otherwise, either the pump radiation would outshine the signal or the signal radiation would be filtered in addition to the pump. In nonlinear interference experiments with terahertz photons involved, atomic gas lasers\cite{56,168,181} at 514.5 nm or single-frequency solid-state lasers\cite{168,182} at 660 nm act as narrow bandwidth sources. Since the phase-matching conditions for conversion processes in LiNbO$_3$ are not critical when terahertz photons are involved, the choice of pump wavelength is not as determining as in the MIR. However, this obstructs the possibility to broaden the generated spectra by a clever choice of the pump wavelength (see Section 2.3.1).

As narrow-band filters, either heated gas cells\cite{56,181} with narrowband absorption at the pump wavelength or volume Bragg gratings (VBG)\cite{168,182} are applied. VBGs are periodical dielectric layers stacked in a way that constructive interference occurs in reflection only for a specific wavelength at a specific angle. This allows to precisely adjust the filters to the laser wavelength by setting the angle, which is a great advantage for experimentalists. At the same time, they provide high attenuation of more than 4 OD and can be positioned one after another. In contrast, for gas cells, the temperature and pressure of the cell must be carefully observed for filtering, in a way that absorption is not broadened to strong.

The crystal material used in the terahertz frequency range for nonlinear interferometers so far is exclusively LiNbO$_3$.\cite{56,168,181,182} This material is already used in many applications for nonlinear generation of terahertz radiation\cite{132,134} and is particularly suitable due to its comparatively high nonlinear coefficient and easy handling. However, it comes with major disadvantages for nonlinear interferometers. On the one hand, it shows a comparatively high absorption in the terahertz frequency range of more than 20 cm$^{-1}$.\cite{183} Thus, a large part of the generated terahertz photons is absorbed in the crystal. On the other hand, the material has a high refractive index of more than five in the terahertz frequency range.\cite{183} As a result, terahertz photons are either Fresnel reflected at the surface or totally reflected already when generated at small angles to the pump radiation. Combined, a large part of the generated terahertz photons does not even leave the crystal, since they are either absorbed or reflected. In addition, terahertz photons leaving the crystal experience large emission angles due to the high refractive index of LiNbO$_3$ in the terahertz frequency range.

Combined, all these hurdles already represent major challenges for the observation of SPDC with terahertz photons involved. Therefore, the observation of nonlinear generation by SPDC or parametric conversion in the terahertz range is already part of the investigations.\cite{54,57,184,188} Due to the generation of down-converted as well as up-converted signal light by conversion of thermal terahertz radiation, the method of crossed dispersion is used for detection (see Figure 6a).

As in the MIR spectral range, various nonlinear interferometer geometries had been applied for measurements in the terahertz frequency range. The approaches can be distinguished primarily by whether only the crystal material or also external samples are studied. As mentioned above, due to the high refractive index the emission angles of the terahertz radiation outside the crystal are very large and cannot be easily collected. This complicates approaches to the study of external samples and explains why early adaptions are performed to study optical properties of LiNbO$_3$ in the terahertz frequency range.

In a first demonstration Kitaeva et al. measured the frequency dependent absorption of undoped and MgO-doped LiNbO$_3$ in the range of 1–2.8 THz\cite{185} by using Young’s geometry. An example of the measurement is shown in Figure 12a. Using a fit of the measured data, the absorption coefficient at the respective wavelength was calculated showing a good agreement to conventional methods. In a later work, Kuznetsov et al. expanded this measurement principle and measured both parts of the dielectric function over the range of 3 THz for nominally congruent LiNbO$_3$ and MgO:LiNbO$_3$ according to the same principle.\cite{187} The measurement of the real part of the dielectric function in the terahertz frequency range is not based on interference but only on the observed frequency-angular spectrum.\cite{184}

Additionally, Kuznetsov et al. measured the linear properties of LiNbO$_3$ using compact Mach–Zehnder geometry.\cite{181,189} Therefore, the LiNbO$_3$ crystal under test was framed by two LiNbO$_3$ crystals with orthogonal orientation of the optical axis. In Figure 12b,c a part of the frequency–angular spectrum measured between 0 and 3 THz is shown in the case of a single crystal and the nonlinear interferometer, respectively. In case of the interferometer, a suppression of the signal is observable for the marked positions at about 1.5 and 2.5 THz. Although the resolution of the measurement is not equivalent to the resolution of the calculation (shown in Figure 12d), the positions match basically giving access to the dispersion properties of the crystal.

Due to the high refractive index of LiNbO$_3$, in the terahertz frequency range, the approach presented in ref.\cite{181} is limited to materials having a similarly high refractive index in this region. Otherwise, losses caused by the large emission angles would be too strong. Therefore, it is necessary to capture as much of the emitted terahertz radiation as possible to build a nonlinear interferometer with terahertz radiation propagating in free space.

In first demonstrations, Kutas et al. placed a two-inch parabolic mirror early after the nonlinear crystal.\cite{168,182} Nevertheless, only a small part of the generated terahertz radiation was captured. The larger part is emitted at greater angles or does not leave the crystal. For this reason, interference was only observable in the collinear forward regions of the frequency–angular spectrum (see Figure 6e), as the corresponding signal photons are emitted under small angles, too.

In the terahertz frequency range, beam-splitting of signal and idler photons can be achieved in two ways. On the one hand, by using dichroic optics as an indium tin oxide (ITO)-coated glass reflecting the terahertz photons but transmitting signal and pump photons.\cite{168} On the other hand, purely dependent on the angle using a parabolic mirror with a through-hole.\cite{182} The omission of the ITO-coated glass has the advantage that no signal or pump photons are reflected at the glass surface. Although there is also a loss of terahertz photons through the hole in the parabolic mirror,
an improvement in interference visibility is observed. In this case, beam-splitting is performed only via the angular emission.

Nonlinear interference experiments with terahertz photons propagating in free space have been demonstrated only in Michelson geometry so far.\cite{168,182} In a proof-of-concept experiment of layer-thickness measurement of PTFE plates ranging from 0.25 up to 5 mm were estimated.\cite{168} In Figure 13c a good agreement of the interferometric measurement with the actual layer thickness is shown.

As a proof-of-concept experiment of terahertz spectroscopy, the extinction of substances showing well known absorption lines were measured.\cite{182} As samples, α-lactose monohydrate and para-aminobenzoic acid were examined with an accessible bandwidth of more than 100 GHz. The comparison with a conventional time-domain spectroscopy (TDS) measurement showed good agreement for both substances. In Figure 13d the comparison of the different measurement methods is shown. While the TDS system used a focused beam to raster scan the entire sample, the beam in the experiment was collimated and passed through the dashed circle (see Figure 13e).

Compared to interference experiments in the MIR, the visibility of the interference experiments with terahertz radiation coupled out of LiNbO₃ is rather low, less than 1%. This is caused by the large number of generated signal photons whose correlated partners are not picked up by the parabolic mirror as well as the large absorption and Fresnel losses. Remaining associated signal photons only form a background and do not contribute to interference. Due to the low visibility of the interference, measurement times are drastically increased compared to measurements in the MIR.

3.3. Imaging with Extreme Light

Although spectroscopic studies in extreme wavelength ranges already benefit from the use of sources and detectors from the visible, the concept of “imaging with undetected photons” is only fully exploited by imaging techniques. These allow the use of low-cost, high-speed, low-noise multipixel sensors, which are usually not available at the concerned wavelength. In particular, two imaging techniques show great potential. OCT allows insights into the depth of samples, while hyperspectral imaging shows its application potential mainly in microscopy of biological samples. But also in the field of imaging based on correlated biphotons in extreme spectral ranges, the demonstrations so far are limited to the low-frequency range.
3.3.1. Mid-Infrared Frequency Range

**Optical-Coherence Tomography**: OCT is a method to investigate structures below the surface. Best known for its application in ophthalmology, this measurement principle shows a wide application in biology, medicine, and non-destructive testing. In this context, common systems for OCT applications work with photons in the visible or NIR spectral range. However, their application for ceramics, paints, or microstructured materials is hindered by strong scattering. MIR photons, on the other hand, achieve much higher penetration depths for these materials and therefore show high application potential. However, the known limitations of the sources and detectors used in MIR remain, for example, due to the needed cooling. Therefore, there is also great potential for concepts based on nonlinear interferometers with correlated photon pairs with idler photons in the MIR.

A different approach based on correlated biphotons is so-called quantum OCT (QOCT), which is based on the interference of degenerate biphotons in a Hong–Ou–Mandel interferometer. By performing correlation measurements, the spectrally entangled photons allow to access depth information. However, previous demonstrations use only degenerate biphotons at NIR wavelengths (800 nm). The applicability of this technique is still limited by both long measurement times due to coincidence detection and limited wavelength tunability. For a detailed discussion please refer to ref. [194].

Based on nonlinear interferometry with idler photons in the MIR, both time-domain OCT (TD-OCT) and frequency-domain OCT (FD-OCT) have been demonstrated so far. In each case, the idler mirror in a Michelson geometry is replaced by the sample under investigation. For TD-OCT, displacing the reference mirror for pump and signal radiation results in an interferogram directly revealing depth information. In case of FD-OCT, the reference mirror remains fixed and the spectral interference recorded by a spectrometer is transferred after data processing via a Fourier transform to obtain depth information. By eliminating the scanning mirror, FD-OCT offers significant advantages in terms of speed and mechanical stability.

The first demonstration of TD-OCT in MIR was performed by Paterova et al. with a similar setup as in ref. [163]. However, to allow for larger translations, pump and signal as well as idler radiation were directly collimated after separation. In addition, the signal is recorded by a single-pixel detector without passing a monochromator. Using a PPLN crystal with regions of different poling period and two different pump sources (at 532 and 488 nm), experiments were realized at several idler wavelengths in the NIR (1543, 2140, and 2504 nm) and MIR (3011 nm). At the MIR wavelength, the optical thickness of a silicon window was determined with an axial resolution of 105 μm (see Figure 14a). Knowing the distances and visibilities of the individual reflections, both the reflection coefficient and refractive index of the sample were calculated with deviations of less than 5% from the literature values.

Compared to measurements carried out in the NIR, the achieved axial resolution is poorer so far. In measurements with idler photons at 1543 nm a resolution of about 41 μm was achieved. In addition, significantly larger visibilities of the observed interference of up to 81% are observed for idler photons at 1543 nm compared to 18% at 3011 nm, due to the used components. Therefore, for further demonstrations, as the determination of birefringence of a sample and raster imaging through a substrate opaque in the visible, idler photons in the NIR were used exclusively. For imaging, an additional short-focal lens placed in the idler beam allowed a lateral resolution of about 50 μm.

The lateral resolution can be further improved by using even shorter focal lengths. On the other hand, axial resolution depends on the used bandwidth and can therefore be significantly improved by generating broadband biphoton spectra. Providing such an extremely broad spectrum, Vanselow et al. demonstrated FD-OCT in the MIR with measurements achieving an axial resolution of 10 μm. Therefore, broadband collinear phase matching was realized in ppKTP at a pump wavelength of 660 nm with idler photons in the range between 3.3 and 4.2 μm (see Section 2.3.1).

In addition, this demonstration showed the great applicability of MIR-OCT in the future by investigating strongly scattering materials and therefore show high application potential. How-
samples in the NIR. A 2D section and a 3D reconstruction of structured samples of alumina ceramics with thicknesses of 900 μm were obtained, using 8 ms of integration time. By focusing on the sample, a lateral resolution of 14 μm was achieved. Additionally, the demonstrated method was compared to conventional NIR-OCT systems at same integration times. The penetration depth of 330 μm achieved in the MIR could not be realized for any of the conventional systems, as can be seen in Figure 14b. As a further application scenario, oil paint covered by a varnish layer on an aluminum foil substrate was also measured, whereby at least the substrate and the air–varnish boundary were resolved.

In comparison, current approaches of OCT in the MIR allowing detection in the visible by up-conversion, only require 3 ms integration time to achieve similar resolutions with higher signal-to-noise ratio. However, for these approaches the power the sample is exposed to is significantly higher than 90 pW used in nonlinear interferometer demonstrations. Although these intensities are not important for material characterization of ceramic samples, this clearly shows great potential with respect to the study of living samples. However, other spectral ranges like the UV illuminate with even higher photon energies and therefore could benefit even more from this measurement principle.

**Microscopy:** The spectroscopic imaging of samples in the MIR is of broad scientific and technical interest, showing major applications in microscopy of biological samples and medicine. Specific signatures of molecules give insight into the study of living samples. However, both absorption and phase of the interference are used as a contrast mechanism for imaging. Therefore, methods based on biphotons show their great potential, allowing the use of sources and detectors in the visible spectral range and only generating low radiation powers at the same time. Especially for the investigation of fragile living cells this is a great advantage.

In contrast to the first demonstration of imaging based on correlated biphotons realizations of imaging in the MIR have been exclusively performed in Michelson geometry. The sample is used either itself, or deposited on a reflective substrate, as the end mirror of the idler arm. Analogous to spectroscopy and OCT, visibility and phase of the interference are used as a contrast mechanism for imaging. However, both absorption and destructive interference can be considered causing dark areas for a single image. To distinguish between these effects, the sample is finely translated in longitudinal (axial) direction, giving access to an interferogram and allowing to retrieve visibility and phase for each individual pixel. In addition, displaying the visibility increases the effective field of view (FoV), since its distribution is much flatter than the intensity distribution of the illumination spot itself.

The most important quantities for characterizing an imaging system are the observed FoV and the achieved resolution. Primarily, these quantities are set by the focal length of the collimating lens used for the idler radiation. To realize microscopy, an additional magnification system (telescope) is introduced in the idler beam path after collimation. It can either be inserted exclusively in the idler arm of the nonlinear interferometer or in both. Starting from a system with a FoV of 9100 μm and a resolution of 322 μm, a FoV of 819 μm with a resolution of 35 μm was realized by means of a tenfold magnification at an idler wavelength of 3.74 μm. By suitable choice of the magnifying optics the spatial resolution of the method can be adjusted application-specifically. Using a x4, x6.7, and x25 magnification spatial resolutions of 78, 50, and 17 μm were achieved at an idler wavelength of 3 μm, as can be seen in Figure 15a.

For both realizations the imaging is based on the momentum anti-correlation of the generated biphotons, for which the sample is placed in the Fourier plane of the crystal. However, the generated biphotons show a strong spatial correlation due to the tight position correlation at birth. Therefore, the sample can also be placed in the image plane, which can be advantageous, as recently shown in ref. In addition, this simplifies the optical system since only a single lens needs to be placed in the optical path. Applying a magnification of only a factor of four, a resolution of 9 μm with a FoV of 161 μm has been demonstrated. However, compared to images in the Fourier plane, the homogeneity of the illumination distribution in the image plane is reduced. Since the photon birth zone itself is illuminated, small inhomogeneities of the crystal or the pump mode as well as dust are of great impact and even result in a reduction of the usable visibility.

In order to assign observed features to spectral ranges, it is necessary to narrow down the observable spectral range. Tuning of the wavelength range plays a major role especially in biological imaging or material characterization. If this examination is performed at different frequencies, the result is a spectroscopic mapping called hyperspectral imaging. This can be realized in several ways. The extension to hyperspectral imaging is particularly straightforward when using broadband biphoton pairs by narrowband filtering of the signal radiation before detection. Using a tunable interference filter with a 3.5-nm bandwidth, spectral selection of ≈85 nm in the range of 3.4–4.3 μm was realized. In addition, the tuning of the wavelength can be done directly by changing the phase-matching conditions via the crystal parameters (poling period, crystal temperature). A first demonstration demonstrates idler photons in the range of 2.8–3.4 μm with a spectral selection of 43 nm. In each case, the spectral resolution results from the bandwidth of the idler photons and thus can be improved by more narrow band filtering or using longer crystals.

In addition, the first application-oriented proof-of-concept experiments have already been realized for microscopy with biphotons in the MIR. On the one hand, Paterova et al. investigated a microfabricated sample of different chemical composition with a resolution of 50 μm (x6.7 magnification) determining the absorption for different idler wavelengths in the range from 2.75 to 3.35 μm. While the sample showed no variance in its optical properties, a significant difference was observed in the MIR for different wavelength ranges. Here, the measured absorption agreed within 4% with the data from a commercial FTIR. Additionally, Kviatkovsky et al. applied the method for investigation of wet biological samples (see Figure 15b). With a resolution of either 35 μm or 9 μm the absorption and phase of a sliced mouse heart was determined for idler wavelengths with a bandwidth of 85 nm around 3.74 μm. Also in this case, structures hidden for conventional visible light microscopy were observed.

In the future, this method has the potential to be integrated into existing microscopes as a supplement. Its great advantage is the low radiation power to which the sample is exposed to. For example, in ref. a radiation power of only 20 pW is given. The obtained measurement times are comparable to those
of commercial systems but still suffer from the necessary variation of the idler arm. In the future, a simple acceleration could be achieved by methods reconstructing the interference by approaching only a few mirror positions.

### 3.3.2. Terahertz Frequency Range

To the best of our knowledge, no imaging based on nonlinear interferometry has been performed in the terahertz frequency range so far. Recently, Kwiatkowsky et al. investigated the theoretical potential of their microscopy technique in the MIR for larger idler wavelengths in the far IR or even in the terahertz frequency range. Since for larger wavelengths the number of resolvable modes decreases drastically, good spatial resolution is challenging to achieve in these extreme spectral regions. To maximize the number of spatial modes, either the pump beam diameter can be increased or the crystal length of the nonlinear material can be shortened. However, in either case the total number of detectable signal photons per angle decreases, leading to even higher measurement times in addition to measurement times due to the high absorption and the high refractive index of LiNbO₃.

Nevertheless, it can be assumed that quantum-inspired measurement techniques based on frequency conversion with pulsed sources could overcome these limitations in the future, since much higher conversion efficiencies can be achieved.

## 4. Outlook

With the high dynamics of the field of quantum sensing with photons in extreme spectral ranges, the barriers will further be pushed. For example, by development of even narrower filters, the transfer to the sub-terahertz or even gigahertz frequency range is within reach. Further, imaging with undetected photons in the terahertz frequency range is not prohibited by physical laws and will therefore most probably be demonstrated in the next years. On the other extreme of the spectrum, first nonlinear interferometers will certainly be realized in the future. Especially in the UV range, all prerequisites are met and experiments in the terahertz frequency range show that nonlinear interferometry can also be performed with high absorption. But also in the X-ray range first implementations are imminent, just like in other frequency ranges ghost imaging experiments could be preceded by nonlinear interferometers. However, the development in all spectral ranges also depends on the available nonlinear materials. If great leaps are achieved for these in terms of transparency ranges, nonlinear coefficients or handling, the same leaps can be expected for the corresponding nonlinear interferometers.

Considering specific applications, the benefits and drawbacks of purely quantum nonlinear interferometers have to be considered. If suitable sources of the interacting photon frequency range are available, seeding of nonlinear interferometers can drastically increase count rates and therefore performance. Not only SPDC contributes to the signal formation, but also photons originating from induced frequency conversion due to the use of a seed source. Therefore, seeding is not limited to be applied to the idler but also improves count rates by application to the signal without imposing much more phototoxic stress on the sample. Initial demonstrations of coherently seeded nonlinear interferometers already show great potential for application and promise higher sensitivities.

Alternatively, the return to classical concepts inspired by quantum optics as nonlinear frequency conversion in combination with phase-retrieving interferometry, can offer advantages in terms of measurement speed. The transfer of photon properties of extreme frequency ranges to the visible can also meet the understanding of electro-optical sampling. Even if the phase and amplitude is not linearly transferred, the photon properties for example, the terahertz range can be transferred to the Si-compatible region and the benefits of 2D detection can be utilized. Therefore, quantum-optical concepts are not the only way to transfer photons properties across the electromagnetic spectrum to regions of good detector availability. Without doubt, for investigation of photon-sensitive samples, for example, in biology or chemistry, quantum-optical concepts provide the benefit of low photon fluxes. As the long hoped-for benefit of a super-resolution has been disproved to be provided simply by the wavelength spread between signal and idler photons, a resolution advantage cannot be gained from SPDC-based nonlinear interferometers.

In the next years, the quantum-optical concepts with extreme light and their potential have to compete with well-established techniques, which themselves are developing further as well.
Depending on the demands of the considered application, the outcome of this competition can be in favor of a classical, quantum-optical, or even mixed technique.

5. Conclusion

In this review article the progress of quantum sensing experiments based on degenerated biphotons with one partner in the extreme wavelength ranges — namely the X-ray, UV, MIR, and terahertz frequency range — is described.

As a key element of these concepts, the development of the generation of such photon pairs by means of SPDC was discussed. In terms of modeling SPDC sources two approaches are commonly used, one based on quantum mechanics and a semi-classical one. Both methods can be applied in different regimes and therefore complement each other well. The application of these approaches can range from cost-effective analytical analysis to full-fledged numerical simulations.

For the state of experimental implementation, the opposing extreme spectral ranges differ significantly. On the high-frequency side, the generation of correlated biphotons is still a major challenge. First demonstrations have already been performed in the X-ray, still having to face current discussion. These experiments suffer strongly from instrumentation as well as the accessible nonlinear materials. Nevertheless, quantum correlation based measurement concepts as ghost imaging were implemented already. The generation of correlated biphotons in the UV range so far is hindered by high absorption and low nonlinear coefficients of the available nonlinear materials.

On the low-frequency side, nonlinear generation has long been a component of classical measurement techniques and therefore provides a great amount of basic knowledge and basic technology. Driven by the requirements of many fields of metrology, the main focus in the MIR is on the realization of broadband SPDC sources, where bandwidths up to 850 cm\(^{-1}\) have been demonstrated recently. In contrast, spontaneous processes in the terahertz spectral range still suffer from absorption or other optical properties of the used nonlinear materials.

The main focus of this review is on the development of measurement concepts in extreme frequency ranges based on “induced coherence”. Although already demonstrated in the early 1990s, the development of this measurement concept has only later regained momentum through “imaging with undetected photons”.

The theory of nonlinear interferometers is well understood on a qualitative level. Explanations for the effects of misalignments in the imaging system and the limitations imposed by the imperfect momentum correlation have been developed. Further various characteristics of the systems can be calculated from analytical models. Numerical models encompassing all the mentioned effects have recently been demonstrated and quantitatively explain experimental results. However, further developments in this area are necessary. Especially simulations of the measured images are important as a basis for tools as they are available for classical optics.

Starting from the generation of non-degenerate biphotons, this measurement principle has been experimentally demonstrated only in the low-frequency range so far. The greatest progress has been achieved for idler photons in the MIR range, were overall wavelengths up to 4.4 μm have been demonstrated. Spectroscopy as well as imaging were realized on the basis of this measurement principle employing a variety of interferometer types, for example, Michelson-type, Mach-Zehnder-type, or compact Mach-Zehnder-type. Especially in the field of spectroscopy, latest demonstrations are on eye level with common methods in terms of resolution and speed. Above all, imaging based on this principle is characterized by the low power the sample is exposed to.

In the terahertz spectral range this concept has been demonstrated exclusively for spectroscopic tasks so far — even in the sub-terahertz range down to 500 GHz. Both the investigation of properties of the nonlinear material itself (addressing bandwidths of more than 1 THz using compact Mach-Zehnder and Young’s geometry) and the linear properties of external samples (with bandwidths of about 100 GHz using a Michelson-type interferometer) have been realized. However, due to low interference visibilities, the measurement times are not yet comparable to current methods and are still in a proof-of-principle status.

Overall, this review showed the great development and the great potential of quantum-based measurement concepts in extreme spectral ranges based on the interference of degenerate biphotons.

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Conflict of Interest

The authors declare no conflict of interest.

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biphoton generation; mid-infrared radiation, nonlinear interferometry, quantum sensing, terahertz radiation, ultraviolet radiation, X-ray radiation

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