Infrared metrology using visible light

Paterova, Anna

2018

Paterova, A. (2018). Infrared metrology using visible light. Doctoral thesis, Nanyang Technological University, Singapore.

<https://hdl.handle.net/10356/103485>

<https://doi.org/10.32657/10220/47387>

INFRARED METROLOGY USING VISIBLE LIGHT

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A thesis submitted to the Nanyang Technological University
in partial fulfillment of the requirement for the degree of

Doctor of Philosophy

2018

Acknowledgements

Working on this thesis was a great journey. Looking back to all these years of my Ph.D. I would like to thank many people for their invaluable support and suggestions.

I would like to express my deepest gratitude to my scientific adviser Dr. Leonid A. Krivitskiy, who always set high goals giving me an opportunity to grow as an independent and professional scientist. Discussions with him were always helpful and fostering the creative thinking.

I would like to also thank my supervisor Prof. Cesare Soci. Despite the short period of work together, comments and suggestions given by him were always constructive and helpful.

I would like to thank all the team members of our research group and especially those, who took direct part in the project, which results were included in this work. Big thanks to Dr. Dmitry A. Kalashnikov for his guidance and transfer of knowledge and skills at the beginning of my Ph.D. Thanks to Dr. Hongzhi Yang for the help with the automation of the experimental setups and with data acquisition. Thanks to Dr. Chengwu An for his always useful comments and remarks on the conducting of experiments.

I would like to thank all the Thesis Advisory Committee members for the numerous discussions, which importance is difficult to overestimate. Thanks to Prof. Rainer Dumke, Prof. Berthold-Georg Englert, Dr. Guillaume Vienne, Prof. Gagik Gurzadyan for their suggestions, which allowed to take a new look into the existing problems.

I would also like to acknowledge my thesis defense panel and thesis reviewers for their contribution to make this work better.

Thank you to Agency for Science, Technology and Research (A*STAR) and A*STAR Graduate Academy (A*GA) for providing me the Singapore International Graduate Award scholarship (SINGA) and all the laboratory facilities for the Ph.D. research.

Thank you to Nanyang Technological University (NTU) for giving me the opportunity to study at the one of the top universities in the world and to get the Ph.D. degree. Thank you to NTU Graduate Program Office (GPO) as well for their guidance over the Ph.D.

Many thanks to my university friends, Shilpa Manandhar, Vasilisa Mishuhina, Julia Gusakova and Ayana Aspembitova, who were always with me to support and encourage.

I am immensely grateful to my family who always supported me and believed in me. Without your love and patience it would be much harder to accomplish this journey.

Thank you too, my dear reader, I hope you will find my work interesting and will gain a new knowledge about the field of the Infrared metrology! ☺

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Abstract

Infrared (IR) spectroscopy and imaging are important tools in metrology, including material analysis, sensing and characterization. Although conventional methods of IR metrology are available, they face challenges related to the high cost and low efficiency of IR light sources and photodetectors. These challenges can be addressed by circumventing the need for IR-range components using tools and techniques of quantum optics.

In this thesis, we describe a technique for IR measurements, which requires using only accessible light sources and detectors operating in the visible range. Our technique is based on the nonlinear interference of correlated photon pairs generated via spontaneous parametric down conversion (SPDC). In SPDC, a photon from the pump laser generates a pair of correlated photons within a nonlinear crystal. One photon of the pair can be generated in the visible range, and the other in the IR range.

Two identical nonlinear crystals pumped by the same laser form the nonlinear interferometer, where interference of the visible SPDC photons can be observed. The interference pattern of the visible photons depends on amplitudes and phases of the IR photons, which are used to probe the properties of the medium under study. Hence, by observing the interference of the visible photons we infer the IR properties of the media, without direct measurements of IR light.

We applied the nonlinear interference method to realize the IR spectroscopy, IR optical coherence tomography (OCT), IR imaging and IR polarimetry. In the IR spec-

troscopy experiment, we simultaneously and independently measured both refractive index and absorption coefficient of various samples in the IR range, while only detecting photons in the visible range. In IR OCT and IR imaging schemes, we measured the reflectivity of the interfaces of the sample, performed 3D raster and wide field imaging through opaque (in the visible range) materials. In the IR polarimetry, we measured the birefringence of the samples. We studied glass, organic polymer, Silicon and CO₂ gas, but our method can be applied to a broad variety of samples.

Our method is widely tunable and has high precision and accuracy over mid-IR wavelengths. This work paves the way for further practical adoption of the method in the field of IR metrology, without the need for IR grade detectors and sources.

Chapter 1

Introduction

1.1 Motivation

Metrology is defined as the science of measurements, and the development of new measurement instruments and techniques is essential to the continued advances of this field. This work focuses on infrared (IR) metrology, which commonly includes the techniques of spectroscopy, optical coherence tomography (OCT) and imaging at IR wavelengths.

This wavelength range is of important interest, as it is where many molecules have spectral fingerprints related to their vibrational and rotational bonds. Consequently, IR spectroscopy [1], IR imaging [2], IR ellipsometry [3] and IR refractometry [4] are important tools in material research and characterization. Conventionally, the IR fingerprint region is defined in the mid-IR range from 2.5 μm to 25 μm .

Since first commercial introduction in 1940, IR spectroscopy has undergone significant development with improved instruments, including the development of dispersive elements and interferometry schemes [1]. The representative IR spectroscopy techniques are dispersion spectroscopy and Fourier Transform Infrared (FTIR) spectroscopy. The FTIR spectroscopy is certainly a predominant technique in the material characteriza-

tion. FTIR devices are available as bench-top and hand-held instruments, and have broad operation range, high spectral resolution and fast readout [5].

Another metrology method is IR imaging [2], which allows analysis of the spatial distribution of molecules or chemicals in a sample. FTIR imaging, which is also well developed, can be used to detect defects and study the composition of various samples such as polymers [6, 7], biological samples [8, 9], etc.

Optical coherence tomography (OCT) is another method in material characterization [10, 11], based on the collection of reflected light from the sample to construct its image. OCT finds broad applications in the characterization of biological samples [12]. It is widely used in ophthalmology [13, 14] for the analysis of the retina and optic disc in eyes, and have recently been developed for cardiology [15, 16].

The resolution and the signal-to-noise ratio of OCT are strongly dependent on the scattering and absorption of the sample. Most commercially available OCT systems operate in the near-infrared (NIR) range, typically in the 850-930 nm band, O- (1260-1360 nm), and C- (1530-1565 nm) bands, which is a compromise between scattering in bio-tissues and absorption by water. Indeed, the Rayleigh scattering is inversely proportional to the wavelength ($\propto 1/\lambda^4$), while the absorption in tissues in near-IR grows with the wavelength.

Table 1.1 gives an overview of the performance of common IR techniques. Despite high accuracy and broad operation range of these techniques, there are several outstanding challenges:

1. There is a limited number of broadband IR light sources, which often requires using several separate sources for multi-wavelength applications (e.g. Quartz Tungsten Halogen (0.7 μm to 3.5 μm) with a Silicon Carbide rod (1.7 μm to 25 μm)). Recently, high power low-coherence quantum cascade superluminescent emitters have been introduced [21]. However, one still needs to use several quantum

Method	IR dispersive spec- troscopy	FTIR spec- troscopy	IR Refrac- tometry [17]	Ellipso- metry [18]	OCT and Imaging [19, 20]
Operation range, cm^{-1} (μm)	20000 - 3000 ($\sim 0.5 - 3$)	20000 - 300 ($\sim 0.5 - 33$)	10000 - 715 ($\sim 1-14$)	5000 - 300 ($\sim 2 - 33$)	12500 - 6300 ($\sim 0.8 - 1.6$)
Spectral resolution, cm^{-1}	~ 3	~ 0.25	~ 0.4	~ 1	~ 8
Accuracy of the measurements					
Refractive index	$\times$	$\times$	10^{-7}	10^{-4}	$\times$
Transmission coefficient	$\sim 10^{-3}$	$\sim 10^{-3}$	$\times$	$\times$	$\times$

Table 1.1 Performance of common IR metrology techniques.

- cascade semiconductor lasers for the broadband operation range. Moreover, the system often requires the cryogenic cooling and have high cost.
2. IR light sources often do not have a smooth spectrum, which leads to complicated analysis and algorithms, experimental inaccuracies and artifacts;
 3. Detectors in the IR range have low quantum efficiency. A common problem of measuring the intensity of IR radiation is the presence of significant thermal noise of the environment with a relatively small useful signal. Detectors of IR radiation should be well isolated from the environment and often cryogenically cooled;

4. IR optical components have a higher cost than their visible-range counterparts and they are often subjected to restrictive export control regulations.

All challenges mentioned above can be addressed using the tools and techniques of nonlinear and quantum optics, which allow circumventing the need for IR-range components by detecting the signal in the visible range.

One such nonlinear optical method is up-conversion spectroscopy, where two visible range lasers (one with a fixed frequency, and one with a tunable frequency) are mixed in a nonlinear crystal to produce a difference frequency signal in the IR range [22, 23]. The IR signal is then used to probe the transmission of a sample and is converted back to the visible range using the frequency up-conversion crystal. One major disadvantage of this method is the requirement of sophisticated tunable lasers, optical system and multichannel detection.

An alternative approach uses the interference of photons produced via spontaneous parametric down-conversion (SPDC) [24, 25]. SPDC occurs in a nonlinear crystal, where the pump laser photon splits into a biphoton, which is a pair of correlated photons called signal and idler [26]. By choosing specific phase matching conditions, one photon can be generated at the visible range (signal photon) and another at the IR range (idler photon).

The nonlinear interferometer can be built using two nonlinear crystals, arranged in such a way that down-converted photons from both crystals are superimposed. Although the signal and idler photons have different wavelengths, the interference pattern of the signal photons depends on the phase and transmissivity of the idler photons in the interferometer. Mandel and colleagues first discovered this effect in 1991, which is now known as induced coherence without induced emission [24, 25].

By detecting the interference pattern for the visible photons, it is possible to perform measurements in IR range, without actually detecting IR photons. This approach

is much simpler than up-conversion spectroscopy, because it does not require the use of tunable lasers. The main advantages of the technique based on the nonlinear interference of SPDC photons are:

1. Measurement of IR properties of the medium using only visible optical components, lasers and detectors, which significantly simplifies the design and cost of the technique;
2. Simultaneous and independent measurement of refractive index and absorption coefficient of the medium, which is hard to achieve in conventional instruments;
3. Measurements over a wide wavelength range (from visible to far infrared range) due to the tunability of the SPDC signal, which makes the system suitable to various applications;
4. Ability to measure various types of samples: gases, liquids, solids, nanoparticles, etc;

1.2 Objectives and Scope

In this work, we demonstrate an approach to IR metrology using visible photons based on nonlinear interference. The main goal of this research is to study the nonlinear interference of SPDC photons and adapt it for various schemes of IR metrology. The performance of the technique is expected to be comparable with conventional techniques. Therefore, the main objectives of this thesis are as follows:

1. To review existing schemes, where the nonlinear interference of SPDC photons is used, and study their applications in metrology;

2. To realize the method for IR spectroscopy with accuracies for refractive index and absorption coefficient comparable with the conventional techniques, and to increase the signal-to-noise ratio of the method;
3. To study theoretically and experimentally the new configuration of the nonlinear interferometer to widen the variety of applications of the method;
4. To study theoretically and experimentally the implementation of the method in IR OCT, IR imaging and IR polarimetry measurements having accuracy and resolution comparable with conventional techniques.

The novelty of the work as follows:

1. Demonstration of IR spectroscopy using visible light with accuracies comparable with the conventional techniques;
2. Demonstration of a new method for improving the signal-to-noise ratio of the method and the readout time;
3. Theoretical and experimental consideration of a new type of the nonlinear interferometer based on the Michelson interferometer. Demonstration of various measurement configurations within the same setup, including IR spectroscopy, IR OCT, IR imaging and IR polarimetry.

1.3 Thesis Outline

The rest of the thesis consists of the following chapters:

- **Chapter 2: Background and methodology.** In this chapter, the background of the proposed metrology technique is described. The background includes the theoretical treatments of SPDC and the nonlinear interference of SPDC photons.

This is followed by an overview of existing schemes for observing of the interference of SPDC photons and their applications in metrology.

- **Chapter 3: Infrared spectroscopy using visible light.** In the first part of this chapter, the application of the technique to IR spectroscopy is described. The method is experimentally demonstrated by measuring the refractive index and the absorption coefficient of carbon dioxide (CO₂) gas. The measurement accuracies are comparable with the conventional techniques: up to $\Delta n = 5 \cdot 10^{-6}$ for the refractive index measurements, and up to $\Delta \alpha = 0.02 - 0.2 \text{ cm}^{-1}$ for the absorption coefficient measurements (depending on the gas concentration). In the second part of the chapter, we demonstrate a method for improving the signal-to-noise ratio in the measurements by employing an image processing algorithm. Results presented in this chapter are published in the following papers: *Nature Photonics* **10**, 98–101 (2016), and *Scientific Reports* **7**, 42608 (2017).
- **Chapter 4: Measurement of infrared optical constants using a non-linear Michelson interferometer.** In this chapter, an extension of the IR spectroscopy technique proposed in Chapter 3 is considered. We design a new measurement scheme based on a nonlinear Michelson interferometer, which eliminates some limitations of the previous scheme, including the necessity of a priori knowledge of the properties of the sample in visible range and possibility of damaging the sample by the pump laser. We demonstrate the IR spectroscopy measurements using a glass, a polymer and a Silicon samples. This method is also shown to have high measurement accuracy: $\Delta n = 0.04$ for the refractive index, and $\Delta \alpha = 0.06 \text{ cm}^{-1}$ for the absorption coefficient. Further improvements in accuracy of the technique is described. Results presented in this chapter are published in the *New Journal of Physics* **20**, 043015 (2018).

- **Chapter 5: Infrared optical coherence tomography and infrared imaging.** In this chapter, IR OCT and IR imaging are realized using the interference of SPDC photons. We measure the reflectivity of internal interfaces of the sample, realize raster and wide field imaging through opaque layers and measure sample birefringence. Our methods of IR OCT and IR imaging are shown to have high lateral and axial resolutions, which is comparable with existing commercial systems. The system is also shown to be highly tunable over the IR wavelengths. Results presented in this chapter are published in the *Quantum Science and Technology* **3**, 025008 (2018).
- **Chapter 6: Polarization properties of the nonlinear interference and infrared polarimetry.** In this chapter, IR polarimetry measurements are shown using the interference of SPDC photons. Using nonlinear Michelson interferometer scheme we probe the birefringence of the sample and demonstrate the accuracy of the measurements up to 0.002π .
- **Chapter 7: Conclusion and future work.** This chapter summarizes the results presented in this work and describes future possible improvements of the method.
- **Appendix.** These chapters contain the technical details of the presented work.

Chapter 2

Background and methodology

2.1 Theory of spontaneous parametric down conversion (SPDC)

The proposed method is based on nonlinear interference of correlated photon pairs generated via SPDC [26, 27]. SPDC is produced in a medium with a quadratic susceptibility $\chi^{(2)}$, where the pump photon generates a pair of strongly correlated photons (or a biphoton) [28] (see Figure 2.1 (a)). The generated photons are usually named signal and idler, where the signal photon has the higher energy. Due to energy and momentum conservation requirements, the frequencies ω and the wavevectors $\mathbf{k}$ of the down-converted photons are given by [26]:

$$\begin{cases} \hbar\omega_p = \hbar\omega_s + \hbar\omega_i \\ \Delta\mathbf{k} = \mathbf{k}_p - \mathbf{k}_s - \mathbf{k}_i \end{cases} \quad (2.1)$$

where indices p, s, i correspond to pump, signal and idler photons, respectively, and $\Delta\mathbf{k}$ is the wavevector mismatch. (In this thesis, bold font indicates vectors).

Fig. 2.1 (a) The process of SPDC in a nonlinear medium with nonzero second order susceptibility $\chi^{(2)}$. Green, orange and red arrows show pump $\mathbf{k}_p$, signal $\mathbf{k}_s$ and idler $\mathbf{k}_i$ wavevectors, respectively. Axis z is parallel to the propagation direction of the pump wavevector. (b) The image of degenerate SPDC photons in the transverse (x, y) plane obtained by a color sensitive CCD camera. The central spot in (b) corresponds to the signal with near-IR wavelengths, which are invisible for the CCD camera.

Figure 2.1 (a) schematically describes the process of SPDC and Figure 2.1 (b) shows the image of the SPDC in the transverse (x, y) plane (perpendicular to the pump propagation direction) detected by focusing into the CCD camera. SPDC photons have the symmetry along the z axis (along the pump propagation direction) and propagate in a cone with the origin inside the crystal [29]. Depending on energy conservation and phase matching conditions given by Equation (2.1), SPDC photons are generated with different wavelengths, which are demonstrated in Figure 2.1 (b). The wavelengths can be tuned by rotating the optical axis of the crystal.

Now, let us consider the mathematical treatment of the SPDC phenomenon. SPDC is based on scattering of pump photons by zero-point fluctuations of the vacuum [26]. Let $E_p(\mathbf{r}, t)$ and $E_i(\mathbf{r}, t)$ describe the electric fields of the pump and idler at the point $\mathbf{r}$ in the crystal, respectively. Then in the point $\mathbf{r}$ the polarization P is [26, 30]:

$$P(\mathbf{r}, t) = \chi^{(2)}(\mathbf{r})E_p(\mathbf{r}, t)E_i(\mathbf{r}, t), \quad (2.2)$$

where $\chi^{(2)}(\mathbf{r})$ is the second order susceptibility.

By presenting the electric field as a sum of negative "(-)" and positive "(+)" frequency parts $E(\mathbf{r}, t) = E^{(-)}(\mathbf{r}, t) + E^{(+)}(\mathbf{r}, t)$ and using Equations (2.1) and (2.2), the interaction Hamiltonian for this process is given by [27, 30]:

$$H_{int}(t) = \frac{1}{2} \int_v d\mathbf{r} \chi^{(2)}(\mathbf{r}) E_p^{(-)}(\mathbf{r}, t) E_s^{(+)}(\mathbf{r}, t) E_i^{(+)}(\mathbf{r}, t) + H.C., \quad (2.3)$$

where v is the interaction volume in the crystal, $H.C.$ is the Hermitian conjugate.

Let us take into account the quantization of the electric field and switch to the quantum description, where electric fields $E(r, t)$ become field operators $\hat{E}(r, t)$ [31, 32]:

$$\hat{E}_s^{(+)}(\mathbf{r}, t) \propto \frac{1}{\sqrt{\nu}} \sum_{\mathbf{k}_s, \sigma_s} \mathbf{e}_{\mathbf{k}_s, \sigma_s} a_{\mathbf{k}_s, \sigma_s} \exp[i(\mathbf{k}_s \mathbf{r} - \omega_s t)], \quad (2.4)$$

where ν is the quantization volume, $\mathbf{e}_{\mathbf{k}, \sigma}$ is the two-dimensional polarization vector, $a_{\mathbf{k}_s, \sigma_s}$ is the photon annihilation operator [33]. The summation σ_s accounts the orthogonal components of a two-dimensional polarization vector, the summation $\mathbf{k}_s$ numbers all possible wavevectors. Similar expression is true for $\hat{E}_i^{(+)}(\mathbf{r}, t)$.

Let us assume the pump wave is given by $E_p(\mathbf{r}, t) = \mathcal{E}(\mathbf{r}) \exp[i(\mathbf{k}_p \mathbf{r} - \omega_p t)]$, where $\mathcal{E}(\mathbf{r})$ is the amplitude of the electric field. Then, from Equations (2.3) and (2.4) the interaction Hamiltonian is defined as [30, 31]:

$$\begin{aligned} \hat{H}_{int}(t) &\propto \frac{1}{2\nu^{2/3}} \sum_{\mathbf{k}_p, \sigma_p} \sum_{\mathbf{k}_s, \sigma_s} \sum_{\mathbf{k}_i, \sigma_i} \exp[i(\omega_s + \omega_i - \omega_p)t] \times \\ &\times \int_v d\mathbf{r} \chi^{(2)}(\mathbf{r}) \mathcal{E}(\mathbf{r}) \exp[-i(\mathbf{k}_s + \mathbf{k}_i - \mathbf{k}_p) \mathbf{r}] a_{\mathbf{k}_s, \sigma_s}^+ a_{\mathbf{k}_i, \sigma_i}^+ + H.C., \end{aligned} \quad (2.5)$$

where $a_{\mathbf{k}_{s/i}, \sigma_{s/i}}^+$ denotes the photon creation operator for the signal/idler photons with wavevector $\mathbf{k}_{s/i}$ and polarization $\sigma_{s/i}$ [33].

The state vector of the SPDC photons $|\psi(t)\rangle$ can be given by the time evolution operator $\hat{U}(t)$:

$$|\psi(t)\rangle = \hat{U}(t) |vac\rangle, \quad (2.6)$$

where $|vac\rangle$ is the initial vacuum state (zero-point fluctuations) at $t = 0$. If the pump field is sufficiently weak, the time evolution operator can be written as:

$$\hat{U}(t) = \exp\left(\frac{1}{i\hbar} \int_0^t dt' \hat{H}(t')\right) = 1 + \int_0^t dt' \hat{H}(t') + \dots, \quad (2.7)$$

where t' is the time variable, t is the interaction time, which is assumed to be small.

Then, according to the first order perturbation theory [34, 35] and using Equations (2.6) and (2.7) the state vector of the SPDC photons in Equation (2.5) is given by:

$$|\psi\rangle = |vac\rangle + \frac{1}{2\nu^{2/3}} \sum_{\mathbf{k}_p, \sigma_p} \sum_{\mathbf{k}_s, \sigma_s} \sum_{\mathbf{k}_i, \sigma_i} F(\mathbf{k}_s, \mathbf{k}_i) a_{\mathbf{k}_s, \sigma_s}^+ a_{\mathbf{k}_i, \sigma_i}^+ |vac\rangle, \quad (2.8)$$

where the two-photon amplitude $F(\mathbf{k}_s, \mathbf{k}_i)$ has the following form:

$$F(\mathbf{k}_s, \mathbf{k}_i) \propto \int_0^t dt' \int_v d\mathbf{r} \chi^{(2)}(\mathbf{r}) \mathcal{E}(\mathbf{r}) \exp[i(\mathbf{k}_p \mathbf{r} - \omega_p t')] \exp[i\Delta\mathbf{k}(\mathbf{k}_s, \mathbf{k}_i) \mathbf{r}] \exp[i(\omega_s + \omega_i - \omega_p) t']. \quad (2.9)$$

We work with the Cartesian coordinate system, where $d\mathbf{r} = dx dy dz$, and make the following assumptions:

1. The pump is a monochromatic plane wave, so that the integral over the time interval t' is reduced to a delta function as $\int_0^t dt' \dots = 2\pi\delta(\omega_s + \omega_i - \omega_p)$ [27], and the summation in Equation (2.8) over the pump photon wavevectors can be neglected;

2. The crystal is uniform ($\chi^{(2)} = \text{const}$), infinite in the transverse directions x and y ; and thin in the longitudinal direction z (so that the walk-off effect of the crystal can be ignored);
3. Small scattering angles of the SPDC photons.

Then, Equation (2.9) takes the following form [36]:

$$\begin{aligned} F(k_s, k_i) &\propto \mathcal{E}(k_{s\perp}, k_{i\perp}) \int_0^L dz \exp[i\Delta k(k_{s\parallel}, k_{i\parallel})z] \\ &= \mathcal{E}(k_{s\perp}, k_{i\perp}) L \text{sinc}\left(\Delta k(k_{s\parallel}, k_{i\parallel}) \frac{L}{2}\right), \end{aligned} \quad (2.10)$$

where L is the crystal length, and $\parallel, \perp$ indices show parallel and perpendicular components of wavevectors relative to z axis, respectively (see Figure 2.2). $F(k_s, k_i)$ defines the angular-frequency spectrum of the two-photon field. In the general case, the angular spectrum of the SPDC is determined by the interplay between the divergence of the pump beam and the phase-matching conditions in the crystal [36–41].

Figure 2.2 (a) shows the SPDC scattering angles $\theta_{s,i}$. Here θ_p is the crystal phase matching angle (angle between pump wavevector and optical axis of the nonlinear crystal) [42]. Figure 2.2 (b) schematically illustrates the phase matching condition of SPDC, where parallel components of the wavevectors in Figure 2.2 (b) define the longitudinal term of the SPDC effect. Figure 2.2 illustrates that each angle of the SPDC is characterized by different wavelengths, which is evidenced from Figure 2.1 (b).

In Equation (2.10) the phase mismatch Δk has only parallel components [27]. Therefore, the phase matching condition is given by the following equations (see Figure 2.2 (b)):

$$\begin{cases} \Delta k = k_p - k_{s\parallel} - k_{i\parallel} = k_p - k_s \cos \theta_s - k_i \cos \theta_i \\ 0 = k_{s\perp} - k_{i\perp} = k_s \sin \theta_s - k_i \sin \theta_i \end{cases} \quad (2.11)$$

Fig. 2.2 (a) Scheme of the SPDC process in a nonlinear crystal showing scattering angles $\theta_{s,i}$. Here θ_p is the phase matching angle of the crystal (optical axis of the crystal is indicated as a dashed-dot line). Green, orange and red arrows indicate pump, signal and idler photons, respectively. (b) The phase matching condition for pump $\mathbf{k}_p$, signal $\mathbf{k}_s$, idler $\mathbf{k}_i$ wavevectors and their projections (dashed lines) into z axis (parallel component) and y/x axis (traverse component).

where $\theta_{s,i}$ are external (outside of the crystal) angles between pump wavevector and signal/idler wavevectors.

The two-photon amplitude $F(k_s, k_i)$ of SPDC can be simplified into a function of two parameters (ω_s, θ_s) using relations (2.1) and (2.11):

$$F(\omega_s, \theta_s) \propto L \operatorname{sinc}\left(\Delta k(\omega_s, \theta_s) \frac{L}{2}\right), \quad (2.12)$$

$$\Delta k(\omega_s, \theta_s) = \frac{n_p(\theta_p)\omega_p}{c} - \frac{n_s\omega_s}{c} \sqrt{1 - \left(\frac{\sin \theta_s}{n_s}\right)^2} - \frac{n_i(\omega_p - \omega_s)}{c} \sqrt{1 - \left(\frac{\omega_s \sin \theta_s}{n_i(\omega_p - \omega_s)}\right)^2}, \quad (2.13)$$

where $n_{p,s,i}$ are the refractive indices of the crystal at the pump, signal and idler photon wavelengths, respectively. Equation (2.13) shows that the phase mismatch can be controlled depending on the phase matching angle of θ_p with respect to the pump wavevector $\mathbf{k}_p$. (Equations (2.12) and (2.13) also can be rewritten in terms of wavelength and angular variables).

Depending on polarizations of the pump and the generated SPDC photons, type-0: $e \rightarrow e + e$, type-I: $e \rightarrow o + o$, and type-II: $e \rightarrow e + o$ phase matching conditions can be achieved, where e and o are an extraordinary and an ordinary waves in the crystal, respectively. Figure 2.3 shows the wavelength-angular spectra of the type-I SPDC

photons for different phase matching angles θ_p of the LiNbO₃ crystal with $L = 0.5$ mm length using a pump with $\lambda_p = 532$ nm wavelength. Figure 2.3 (a) shows the degenerate regime of the SPDC, where $\lambda_s = \lambda_i = 2\lambda_p$ at zero angle (along z axis in Figure 2.2 (a)). Figure 2.3 (b) shows the frequency non degenerate and collinear regime of the SPDC, where wavelength of the signal photons is blue-shifted and the wavelength of the idler photons is red-shifted.

By choosing the phase matching angle of the crystal θ_p it is possible to generate SPDC photons in a strongly non-degenerate regime, when the signal photon wavelength is in the visible range and the idler photon wavelength is in the IR range. Alternatively, the phase matching can be controlled by heating the crystal [43], by applying electric fields [44], or by using specialized crystals with periodic poling [45, 46].

For a single spatial mode, it is possible to rewrite Equation (2.12) in terms of frequency mismatches Ω as [47, 48]:

$$\begin{cases} \omega_s = \omega_{s0} - \Omega \\ \omega_i = \omega_{i0} + \Omega \end{cases} \quad (2.14)$$

where ω_{s0}, ω_{i0} are central frequencies of the signal and idler photons, respectively. Then, Equation (2.12) is given by:

$$F(\Omega) \propto \int_0^L dz \exp(i\Delta k(\Omega)z) \propto L \operatorname{sinc}\left(\Delta k(\Omega)\frac{L}{2}\right). \quad (2.15)$$

The first-order correlation function of the SPDC photons is given by [49, 50]:

$$G^{(1)}(\Delta t) \propto \int |F(\Omega)|^2 \cos(\Omega \Delta t) d\Omega, \quad (2.16)$$

where Δt is the time delay between signal and idler photons.

Fig. 2.3 Simulation of SPDC wavelength-angular spectrum from LiNbO₃ crystal with $L = 0.5$ mm length using a pump with $\lambda_p = 532$ nm wavelength. **(a)** Degenerate regime of SPDC photons for $\theta_p = 81.4^\circ$. Its center corresponds to zero angle and degenerate wavelength $\lambda_s = \lambda_i = 2\lambda_p$. **(b)** Non-degenerate regime of SPDC for $\theta_p = 78.0^\circ$. Here SPDC spectrum has two separated branches with $\lambda_s < \lambda_i$.

Equation (2.16) is given by the Wiener–Khinchin theorem [51]. The width of the correlation function Δt_c (or coherence time) is inversely proportional to the width of the frequency spectrum $\Delta\omega_s$: $\Delta t_c \propto \frac{1}{\Delta\omega_s} = \frac{1}{\Delta\Omega}$. These properties play a decisive role in the quantum optical coherence tomography (QOCT) [52–55], where the resolution of the method is defined by the coherence length $l_{coh} = c \Delta t_c$.

2.2 Theory of induced coherence without induced emission

Now let us consider the case of two nonlinear crystals, which are coherently pumped using continuous wave (CW) laser (see Figure 2.4). Let us assume type-0 or type-I phase matching condition, where $\sigma_i = \sigma_s = \sigma$, and the summation over σ may be omitted. From Equation (2.8), the resulting state vector for spatial and frequency modes of the SPDC photons in the interferometer is given by a superposition of state vectors of down-converted photons generated from the first "1" and the second "2"

nonlinear crystal:

$$\begin{aligned}
 |\Psi\rangle = |\psi_1\rangle + |\psi_2\rangle = |vac\rangle + \frac{1}{2\nu^{2/3}} \sum_{\mathbf{k}_s, \mathbf{k}_i} F(\mathbf{k}_s, \mathbf{k}_i) a_{\mathbf{k}_{s1}}^+ a_{\mathbf{k}_{i1}}^+ |vac\rangle + \\
 + \frac{e^{i\phi_p}}{2\nu^{2/3}} \sum_{\mathbf{k}_s, \mathbf{k}_i} F(\mathbf{k}_s, \mathbf{k}_i) a_{\mathbf{k}_{s2}}^+ a_{\mathbf{k}_{i2}}^+ |vac\rangle,
 \end{aligned}
 \quad (2.17)$$

where ϕ_p is the phase acquired by the pump [56, 57].

Let us assume that idler modes $i1$ and $i2$ are fully matched at the output of the crystal "2", i.e. $i1 = i2 = i$. If an object is inserted into the path of idler photons $i1$, the losses induced by the object can be described using a beam splitter model, where a vacuum state is introduced into the empty port of the beam splitter [58] (see inset in Figure 2.4). Thus, the photon creation operator for the idler photons at the second

Fig. 2.4 **Scheme of a nonlinear Mach-Zehnder interferometer.** Two identical nonlinear crystals are pumped by the continues wave (CW) laser, which is divided initially into two paths by the beam splitter BS1. SPDC photons are generated in each crystal. The idler modes from the first ($i1$) and the second ($i2$) crystals are aligned. An object with an amplitude transmission coefficient τ_i is placed in the path of the idler photons generated from the first crystal. The signal photons $s1$ and $s2$ are mixed in the beam splitter BS2. Here and further the green, yellow and red arrows indicate pump, signal and idler wavevectors, respectively. The dashed lines show the trajectories of the photons. The inset schematically illustrates the beam splitter model.

crystal is given by [59]:

$$a_{\mathbf{k}_{i2}} = e^{i\phi'_i}(\tau_i a_{\mathbf{k}_{i1}} + \sqrt{1 - \tau_i^2} a_{\mathbf{k}_{i0}}), \quad (2.18)$$

where $a_{\mathbf{k}_{i0}}$ describes the vacuum field entering from the unused port of the beam splitter, τ_i is the complex amplitude transmission coefficient of idler photons through the medium, ϕ'_i is the phase acquired by the idler photons. Then, the resulting state vector in Equation (2.17) is given by:

$$\begin{aligned} |\Psi\rangle = & |vac\rangle + \frac{1}{2\nu^{2/3}} \sum_{\mathbf{k}_s, \mathbf{k}_i} F(\mathbf{k}_s, \mathbf{k}_i) a_{\mathbf{k}_{s1}}^+ a_{\mathbf{k}_{i1}}^+ |vac\rangle + \\ & + \frac{e^{i(\phi_p - \phi'_i)}}{2\nu^{2/3}} \sum_{\mathbf{k}_s, \mathbf{k}_i} F(\mathbf{k}_s, \mathbf{k}_i) a_{\mathbf{k}_{s2}}^+ (\tau_i^* a_{\mathbf{k}_{i1}}^+ + \sqrt{1 - \tau_i^{*2}} a_{\mathbf{k}_{i0}}^+) |vac\rangle. \end{aligned} \quad (2.19)$$

When the signal modes $s1$ and $s2$ are superimposed on a beam splitter, the count rate of signal photons is given by [35, 59]:

$$P_s \propto \langle \Psi | E_s^{(-)}(t) E_s^{(+)}(t) | \Psi \rangle, \quad (2.20)$$

where $E_s^{(+)}(t)$ is the electric field at the detector:

$$E_s^{(+)}(t) \propto \sum_{\omega} a_{s1} e^{-i\omega(t-t_{s1})} + a_{s2} e^{-i\omega(t-t_{s2})}, \quad (2.21)$$

and $t_{s1, s2}$ are travel times of signal photons from the crystal "1" and "2" to the detector, respectively. Using Equation (2.19) and taking into account that $\phi'_i = \omega_i t_{i1}$, where t_{i1} is the travel time of the idler photons $i1$ in the interferometer, we obtain the following equation:

$$\begin{aligned} E_s^{(+)}(t) | \Psi \rangle \propto & \frac{1}{2\nu^{2/3}} \sum_{\mathbf{k}_s, \mathbf{k}_i} F(\mathbf{k}_s, \mathbf{k}_i) \left(e^{-i\omega_s(t-t_{s1})} a_{\mathbf{k}_{i1}}^+ + \right. \\ & \left. + e^{-i\omega_s(t-t_{s2}) + i(\phi_p - \omega_i t_{i1})} (\tau_i^* a_{\mathbf{k}_{i1}}^+ + \sqrt{1 - \tau_i^{*2}} a_{\mathbf{k}_{i0}}^+) \right) |vac\rangle. \end{aligned} \quad (2.22)$$

Then, the count rate of the signal photons is given by:

$$P_s \propto \frac{1}{2\nu^{2/3}} \sum_{\mathbf{k}_s, \mathbf{k}_i} \left| F(\mathbf{k}_s, \mathbf{k}_i) \right|^2 \left| e^{-i\omega_s(t-t_{s1})} + e^{-i\omega_s(t-t_{s2})+i(\phi_p-\omega_i t_{i1})} \tau_i^* \right|^2 + \left| e^{-i\omega_s(t-t_{s2})+i(\phi_p-\omega_i t_{i1})} \sqrt{1-\tau_i^{*2}} \right|^2. \quad (2.23)$$

Assuming that the quantization volume ν is large enough, the summation over wavevectors can be substituted by integrals [30]. Redefining the frequencies in terms of detuning similar to Equation (2.14), the photon count rate is:

$$P_s \propto \int d\Omega \left| F(\Omega) \right|^2 \left(1 - \tau_i^{*2} + \left| e^{-i(\omega_{s0}-\Omega)(t-t_{s1})} + e^{-i(\omega_{s0}-\Omega)(t-t_{s2})+i(\phi_p-(\omega_{i0}+\Omega)t_{i1})} \tau_i^* \right|^2 \right). \quad (2.24)$$

Taking into account the normalization of the two-photon amplitude $\int d\Omega \left| F(\Omega) \right|^2 = 1$, Equation (2.24) takes the form:

$$P_s \propto \left(2 + \left(\tau_i^* e^{i(\phi_p+\omega_{s0}(t_{s2}-t_{s1})-\omega_{i0}t_{i1})} \int d\Omega \left| F(\Omega) \right|^2 e^{-i\Omega(t_{s2}-t_{i1}-t_{s1})} + c.c. \right) \right), \quad (2.25)$$

where *c.c.* is a complex conjugate term.

With $\omega_{s0}(t_{s1} - t_{s2}) = \phi_s$ and $\omega_{i0}t_{i1} = \phi_i$ being the accumulated phases of signal and idler photons in the interferometer, the final expression for the count rate at the detector is given by [24, 25, 60, 61]:

$$P_s(\Delta t) \propto 2 \left(1 + |\tau_i| |\mu(\Delta t)| \cos \left(\phi_p - \phi_s - \phi_i + \arg(\tau_i) + \arg(\mu(\Delta t)) \right) \right), \quad (2.26)$$

$$\mu(\Delta t) = \int |F(\Omega)|^2 \exp(-i\Omega \Delta t) d\Omega.$$

The introduced function $\mu(\Delta t)$ coincides with Equation (2.16) and denotes the normalized first order correlation function of the SPDC [48, 49] with the time delay $\Delta t = t_{i1} + t_{s2} - t_{s1}$ in the interferometer. The correlation function $\mu(\Delta t)$ defines the envelope for the interference fringes (see Figure 2.5). The interference fringes are

observed only within the coherence time of the SPDC Δt_c , which is defined by a full width at half maximum of interference fringes. Since the correlation function $\mu(\Delta t)$ is normalized, the amplitude of the interference pattern is determined by the transmission function τ_i^m .

The interference pattern, given by Equation (2.26), is defined as function of the time delay, and the visibility of the interference fringes is given by the ratio:

$$V = \frac{P_s^{max} - P_s^{min}}{P_s^{max} + P_s^{min}} = |\tau_i| |\mu(\Delta t)|. \quad (2.27)$$

Equation (2.26) is defined for a single spatial mode. However, the SPDC has also a spatial distribution [39]. Due to the divergence of the SPDC the interference pattern has a modulation in the spatial domain and has a ring structure [62]. The modulation of the SPDC in the spatial domain is determined by the arguments under the cosine function in Equation (2.26).

Fig. 2.5 Illustration of the interference fringes in a time domain. The left figure shows how the correlation function $\mu(\Delta t)$ defines the envelope of the interference fringes. The right figure shows the interference pattern, which can be obtained in the experiment. The interference pattern is observed within the coherence time of the SPDC photons Δt_c , when the interferometer arms are balanced. The amplitude of the interference fringes is also determined by the amplitude transmission τ_i^m of the object.

Besides time domain in Equation (2.26), the interference pattern can be determined in the frequency domain: [63, 64]:

$$P_s(\omega_s) \propto |F(\omega_s)|^2 \left(1 + |\tau_i| \cos \left(\phi_p - \phi_s - \phi_i + \text{Arg}(\tau_i) \right) \right), \quad (2.28)$$

where $|F(\omega_s)|^2$ is the spectral density of the SPDC. The interference is observed, when frequency resolution is much narrower than inverse value of optical path difference $1/\Delta t$, which makes the time delay Δt unresolvable.

2.3 Nonlinear interferometers and their applications

It is well known that the interference of two light beams depends on their mutual coherence and the intrinsic indistinguishability of their propagation paths [65]. In this section we consider several interferometric schemes [66], where interference of SPDC photons from two nonlinear crystals is observed, and describe how the mutual coherence is achieved.

2.3.1 Nonlinear Mach-Zehnder interferometer

The superposition of the state vectors of down-converted photons results in interference only when they are mutually coherent [65]. In quantum mechanics the "mutual coherence" refers to indistinguishability of particle trajectories (or "which path" information). Thus, the interference of single photons in the interferometer shown in Figure 2.4 can be observed, when one cannot distinguish the crystal in which SPDC photon pair is generated. The degree of the mutual coherence (or indistinguishability) includes the longitudinal coherence (when interferometer paths are balanced), transverse coherence (the spatial mode overlap of the beams), and the losses in the system between crystals.

The nonlinear interference of SPDC photon pairs was first demonstrated by L. Mandel and colleagues in 1991 [24, 25] and was called the "induced coherence without induced emission" effect. In the experiment, the nonlinear Mach-Zehnder interferometer (see Figure 2.4) was built using a mirror, a beam splitter BS2, two identical nonlinear crystals, where SPDC photons were generated using a CW laser as a pump. The mutual coherence of SPDC photons from two crystals was achieved by aligning idler modes $i1$ and $i2$ to make them indistinguishable and by mixing signal photons on the beam splitter BS2. When the neutral density filter with amplitude transmission function τ_i was inserted into the path of idler photons $i1$, the degree of mutual coherence was determined by the visibility of the interference fringes in Equation (2.27) [67].

In the "induced coherence" experiments the modulation of the signal photon counts on the detector versus the displacement of the beam splitter BS2 was observed [24, 25]. The experiment demonstrated that the visibility of the interference fringes of signal photons also depended on the idler photons (see Equation (2.26)).

In the experiment, the authors achieved a visibility of $V = 30\%$. Such a low visibility can be explained by the imperfect overlap of the SPDC spatial modes [62, 67–70], the non-balanced paths of photons in the interferometer [61], and the losses in the channel of idler photons $i1$.

The effect of the overlap of the spatial modes on the visibility of the interference pattern have been studied as well. In the papers [67, 69], the authors emphasized that SPDC beams have a finite transverse spatial dimension and divergence. Consequently, there are regions, where it is possible to distinguish whether a photon was created in the first or in the second crystal. Thus, the visibility of the interference pattern decreases with increasing the scattering angles. The authors provide a treatment for the visibility of the interference pattern that accounts for the overlap of the spatial modes of the

beams and find that the theoretical visibility decreases from unity to $V = 0.67$ [67]. However, it is possible to restore the degree of coherence in the interferometer by compensating for the divergence of the SPDC photons from the first crystal using lenses [59, 68].

The longitudinal coherence also affects the visibility of the interference. In this case, the degree of indistinguishability is the first order correlation function [60, 61, 71] (see Equation (2.26)). Therefore, the interference fringes can be observed when all optical path lengths in the interferometer are within the coherence length l_{coh} of the SPDC photons. However, the interference can still be observed in the mixing of signal photons, even if the optical path difference through the interferometer exceeds the coherence length of the SPDC photons. When the frequency resolution is much narrower than inverse value of optical path difference $1/\Delta t$ (which means the indistinguishability in the frequency domain), the modulation appears in a frequency domain (in the spectrum of the SPDC) [63] as discussed previously (see Equation 2.28).

The practical relevance of the nonlinear Mach-Zehnder interferometer is shown in the quantum imaging experiment [72], where authors performed IR imaging by detecting visible photons. The imaging was done in a transmission mode using a so-called " $4f$ imaging configuration", where the angular spectrum of the SPDC photons was modified (see Figure 2.6 (a)). The $4f$ imaging configuration includes two identical lenses, where crystals, lenses and the object are put as shown in the inset of Figure 2.6. The same lens configuration was used for the overlap of the signal photons.

This configuration gives two crucial insights:

1. The spatial modes of SPDC photons from the first and second crystals are efficiently overlapped. Therefore, in the CCD camera the interference pattern is recorded as a single spot depending on introduced phases in the interferometer.

2. The object is placed in the focal plane of the first lens, where the k -space (wavevectors) of the SPDC idler photons is imaged. Therefore, each wavevector probes the transmissivity of an object in a transverse (x, y) plane.

Fig. 2.6 Quantum imaging experiment using a nonlinear Mach-Zehnder interferometer. In the experiment the authors performed phase imaging of a fused silica sample, with probing wavelength at 1550 nm, and detected wavelength at 810 nm. (a) The scheme of the experimental setup (b) The fused silica, which was used as an object under study (c) The image at the CCD camera, with different introduced phases in the interferometer. The inset illustrates the $4f$ lens configuration. Figures (b) and (c) are used with the permission from [72].

The authors used a fused silica plate with the shape of a cat as a sample (see Figure 2.6 (b)), where probing idler photons at 1550 nm wavelength acquired an additional phase shift of π . The wide-field phase-image of the sample was detected at the conjugate wavelength of the signal photons at 810 nm in k -space (see Figure 2.6 (c)).

2.3.2 Nonlinear degenerate Mach-Zehnder interferometer

The interference of SPDC photons can also be observed in a degenerate nonlinear Mach-Zehnder interferometer (see Figure 2.7 (a)). In contrast with the previous non-degenerate interferometer, in the degenerate nonlinear Mach-Zehnder interferometer the two crystals are placed sequentially and signal photons $s1$ propagate through the second nonlinear crystal as well. In the literature this interferometer was first considered by D. N. Klyshko [73, 74].

The theory for the interference pattern treatment coincides with the previous case, with the difference in the phase of the signal photons under the cosine function. In this interferometer the phase ϕ_s includes the phase gained inside the second crystal too.

Fig. 2.7 **Scheme of the nonlinear degenerate Mach-Zehnder interferometer.** (a) Two identical nonlinear crystals are placed sequentially in the path of the common pump beam. SPDC photons are generated in each crystal. A sample is placed in the gap between nonlinear crystals. (b) The degenerate nonlinear Mach-Zehnder interferometer is represented as a set of one nonlinear crystal and a mirror. A sample is placed in the gap between the nonlinear crystal and the mirror. In both schemes all three photons propagate through the sample and dashed lines show the trajectories of the photons.

Taking into account Equation (2.13), it is possible to redefine the arguments under the cosine function in Equation (2.26) [75, 76]:

$$\begin{cases} \Phi = \phi_p - \phi_s - \phi_i = \delta + \delta^m \\ \delta = k_p L - k_s L - k_i L = \Delta k L \\ \delta^m = k_p^m L^m - k_s^m L^m - k_i^m L^m = \Delta k^m L^m \end{cases} \quad (2.29)$$

where L^m is the length of the gap between crystals, δ and δ^m determine the phases that the photons acquire inside the crystal and in the gap, respectively. (Here, we use the definition of the phase ϕ as a product of wavevector and an optical path). From Equation (2.29), parameters Δk and Δk^m denote the phase matching condition inside the crystal and in the gap between crystals, respectively.

Therefore, the interference pattern in a frequency-angular domain is given by:

$$P_s(\omega_s, \theta_s) \propto |F(\omega_s, \theta_s)|^2 \left(1 + |\tau_i| \cos(\delta + \delta_m) \right). \quad (2.30)$$

The example of the interference pattern in Equation (2.30) is shown in Figure 2.8. Similarly, as in Figure 2.3, two LiNbO₃ crystals with $L = 0.5$ mm length are used. The crystals are pumped by the CW laser with $\lambda_p = 532$ nm wavelength. The length of the gap between crystals is set to be $L^m = 1$ mm.

In this interferometer configuration, the visibility of the interference pattern depends on the mutual coherence of SPDC photon pairs generated at the first and second nonlinear crystals [77, 78].

The longitudinal coherence varies slowly with the change of the gap length, because of the degeneracy of the interferometer. Since all the three photons (*pump*, *signal*, *idler*) propagate along the same path in the interferometer, the time delay Δt is determined by the difference in propagation times of the signal ($t_s = t_{s1} - t_{s2}$) and idler (t_{i1})

Fig. 2.8 Interference patterns of the SPDC photons in a degenerate Mach-Zehnder interferometer using two LiNbO₃ nonlinear crystals with $L = 0.5$ mm length, $\lambda_p = 532$ nm and an air gap between crystals is $L^m = 1$ mm. **(a)** Degenerate SPDC interference for $\theta_p = 81.4^\circ$. **(b)** Non-degenerate SPDC interference for $\theta_p = 78.0^\circ$.

photons:

$$\Delta t = (n_i - n_s) \frac{L}{c} + (n_i^m - n_s^m) \frac{L^m}{c} \quad (2.31)$$

where $n_{s,i}^m$ are the refractive indices of the medium in the gap at the signal and idler wavelengths, respectively. In case of an air gap between crystals, the change in optical delay is negligibly small.

Moreover, this interferometer is intrinsically stable compared to non-degenerate configuration, because of the spatial degeneracy of the interferometer arms [75]. The phase change due to perturbations in the interferometer (temperature, pressure or environmental fluctuations) is given by [75]:

$$\frac{d\Phi}{dL^m} = \frac{d\delta^m}{dL^m} = 2\pi \left(\frac{n_p^m}{\lambda_p} - \frac{n_s^m \cos \theta_s}{\lambda_s} - \frac{n_i^m \cos \theta_i}{\lambda_i} \right), \quad (2.32)$$

where dL^m is the change of the length of the interferometer gap, n_p^m is the refractive index of the medium in the gap at the pump wavelengths.

Here, the phase inside the crystal δ is constant. From Equation (2.32), the effect of a small displacement of one of the crystals on the intensity distribution of the

interference pattern is negligibly small, assuming small scattering angles $\theta_{s,i}$ and weak dispersion of the object.

Considering the spatial overlap of the SPDC modes, the interference pattern is observable under the assumption that the signal photons do not exceed the limits of the interaction region in the second crystal [78–80]:

$$(2L + L^m) \tan(\theta_s) \ll d, \quad (2.33)$$

where d is the pump beam diameter. In this case, observer cannot distinguish where the SPDC photons were generated ("which-path" information).

This interferometer configuration is easier to align compared to the non-degenerate one, and there is no need for active phase stabilization. An even simpler geometry of the degenerate Mach-Zehnder interferometer with one nonlinear crystal and a mirror is shown in Figure 2.7 (b) [75]. Besides the intrinsic stability, the phase matching is automatically achieved, since all the SPDC photons are generated in the same crystal.

Nonlinear interference of SPDC photons in a nonlinear degenerate Mach-Zehnder interferometer was applied to IR spectroscopy of linear media [75, 76]. In the work of D. Yu. Korystov et al. [76] crystalline quartz was placed in the gap between two crystals and the interference pattern in the wavelength-angular spectrum was observed (similar to Figure 2.8 (b)). Interference fringes were determined by the dispersion characteristics of the quartz sample. Depending on the phase-matching conditions, the signal and idler photons could be generated at the visible and IR range, respectively. Hence, it was possible to measure the dispersion characteristics of the crystalline quartz in the IR range by detecting visible photons. In the experiment the authors achieved an accuracy of the refractive index measurement about $\Delta n = 10^{-2}$. However, the accuracy could be further improved, since the precision of the method mainly depended on the accuracy of determining the angular coordinates.

The interference fringes become curved (see Figure 2.9) [81], if the medium inside the gap exhibits a resonance behavior at the wavelength of idler photons. This hooks are well known as "Rozhdestvenski hooks", which are obtained in an interferometer combined with a grating spectrograph [82–84].

In this case the transmission of idler photons through the gap is determined by the Bouguer law (or the Beer-Lambert law):

$$|\tau_i| = \exp(-\alpha_i^m L^m / 2), \quad (2.34)$$

where α_i^m is the absorption coefficient of the medium introduced in the gap at the idler wavelength.

In the work of D. Yu. Korystov et al. [81] the dispersion characteristics of paraffin oil near a resonance was measured (see Figure 2.9). Figure 2.9 (a) shows the signal

Fig. 2.9 Interference of SPDC photons in a nonlinear degenerate Mach-Zehnder interferometer with the medium in the gap having resonance behavior at the wavelength of idler photons. Wavelength-angular spectrum for (a) a single LiNbO₃ crystal with $L = 440 \mu\text{m}$, (b) two LiNbO₃ crystals with $L^m = 50 \mu\text{m}$, where the interference fringes are determined by the dispersion characteristics of the paraffin oil placed in the gap. The interference fringes present hooks because of the resonance behavior of the medium under study at the wavelength of idler photons. These figures are used with the permission from [81].

for the SPDC measured from one crystal (LiNbO_3 with $L = 440 \mu\text{m}$). Figure 2.9 (b) shows the interference of SPDC photons from two crystals separated by a gap $L^m = 50 \mu\text{m}$, which was filled with paraffin oil. The "hooks" at the interference pattern indicate the presence of the resonance dispersion of the medium at the idler photon wavelength.

Another application of the degenerate Mach-Zehnder interferometer is the measurement of the absorption coefficient using Equation (2.34). In the work of S. P. Kulik et al. the absorption of a thin gold film deposited on a fused silica substrate was measured [77]. However, the measured value of the absorption coefficient differed five-fold from the literature value, which could be explained by experimental inaccuracies.

2.3.3 Nonlinear Michelson interferometer

Another geometry of the interferometer is shown in Figure 2.10. Here, the SPDC photons are split in two different directions and then all the photons are reflected back by mirrors into the same crystal. This configuration forms a nonlinear Michelson interferometer [57].

The visibility of the interference pattern in the nonlinear Michelson interferometer also depends on the spatial overlap of the modes, temporal coherence of SPDC photons [85–88] and losses for the idler photons in the interferometer.

Interference pattern can be observed when the interferometer arms are balanced.

Fig. 2.10 **Scheme of a nonlinear Michelson interferometer.** SPDC field occurs in the forward and backward passes of the pump beam through the nonlinear crystal. The pump beam and the SPDC field are reflected back by mirrors along the same paths (dashed lines show the trajectories of the photons).

This geometry of the interferometer is similar to the degenerate Mach-Zehnder interferometer with one crystal and a mirror. Here, the automatic phase matching alignment of SPDC photon pairs is also achieved. In such an interferometer all photons travel in different paths, and the phase delay can be introduced independently for each photon (*pump*, *signal*, *idler*). However, this also means that the alignment of the interferometer becomes more challenging, and the setup is more prone to instabilities.

The interference of SPDC photons in the nonlinear Michelson interferometer was recently applied for imaging experiment [89], which also included the modification of the angular spectrum of the SPDC photons as demonstrated in the inset of Figure 2.6 [72].

2.3.4 Nonlinear Young's interferometer

A nonlinear Young's interferometer is shown in Figure 2.11 (a). Here, crystals are aligned in the transverse direction [90]. The SPDC photon pairs generated from two crystals can overlap and become indistinguishable in the far-field.

Fig. 2.11 **Schemes of a nonlinear Young's interferometer.** (a) Two nonlinear crystals are aligned in a transverse direction and pumped by a CW laser. (b) One nonlinear crystal is used, which is pumped by a CW laser. The transverse mode of the laser beam is modified using a mask. In both cases, the pump generates SPDC photons propagating through the nonlinear crystals. The interference of the SPDC photons can be observed in the far-field zone.

Instead of using two crystals it is possible to put a double slit in front of the single crystal [78, 80, 91] (see Figure 2.11 (b)). This interferometer configuration also has an intrinsic stability and does not need an active phase control.

Assuming a thin crystal and small scattering angles of the SPDC photons, the double slit can be placed even after the crystal. In this case, it is possible to input multiple slits after the crystal to observe multi-beam interference [78].

The visibility of the interference fringes may be reduced due to absorption inside the nonlinear crystal at the wavelength of the idler photons. Therefore, from the interference fringes of the signal photons it is possible to measure the absorption in the nonlinear crystal at the wavelength of idler photons. In the work of A. V. Burlakov and et al. [91] the absorption in a α -HIO₃ crystal was measured. Recently, the same method was used for the dispersion characteristics measurement for pure LiNbO₃ and LiNbO₃:Mg at the THz frequency range [92].

2.3.5 Interferometers with more than two nonlinear crystals

Besides the interferometer geometries described earlier, there are several recent works proposing new configurations with more than two nonlinear crystals [93, 94]. The interference of SPDC photons from N sequentially placed nonlinear crystals coherently pumped by a common laser beam was first considered by D. N. Klyshko, where he theoretically treated the modulation of the signal photon intensity in a degenerate Mach-Zehnder interferometer with N nonlinear crystals [74]. Using many crystals can enhance the contrast of the nonlinear interference pattern, increasing also the signal-to-noise ratio of the method.

Chapter 3

Infrared spectroscopy using visible light

In this chapter, the scheme of the nonlinear degenerate Mach-Zehnder interferometer is considered (shown in Figure 2.7), where all three photons (pump, signal and idler) travel through the sample of interest. The main advantage of this configuration is that the system is robust to perturbations due to the intrinsic stability of the interferometer.

As discussed in section 2.3, the nonlinear degenerate Mach-Zehnder interferometer has been studied for the measurements of the refractive indices of a crystalline quartz and paraffin oil [76, 81], and the absorption coefficient of a thin gold layer [77]. In these works the refractive index was measured with accuracy $\Delta n = 0.01$, whereas the absorption coefficient measurement differed five-fold from the literature value, which could be a result of several factors.

Here, we extend the technique to perform spectroscopic measurements of CO₂ gas, simultaneously obtaining a highly accurate refractive index and absorption coefficient. We also demonstrate a modification of the technique to make it more cost efficient, less time consuming and with a higher signal-to-noise ratio.

3.1 Infrared spectroscopy of CO₂ gas

Let us consider the degenerate nonlinear Mach-Zehnder interferometer shown in Figure 2.7 (a). The interferometer consists of two similar nonlinear crystals with the length L , which are placed at a distance L^m from each other. The optical axis of the crystals is set to generate the signal photons in the visible range and idler photons in IR range. Here, we consider small angles of the generated SPDC, when the condition (2.33) is fulfilled. In this case the intensity of signal photons is given by (see Equations (2.29), (2.30)):

$$\begin{cases} P_s(\omega_s, \theta_s) \propto \frac{1}{2} \left(\text{sinc}\left(\frac{\delta}{2}\right) \right)^2 \left(1 + |\tau_i| \cos(\delta + \delta^m) \right) \\ \delta = \Delta k L = (k_p - k_s - k_i) L = \left(\frac{2\pi n_p}{\lambda_p} - \frac{2\pi n_s}{\lambda_s} \cos \theta_s - \frac{2\pi n_i}{\lambda_i} \cos \theta_i \right) L \\ \delta^m = \Delta k^m L^m = (k_p^m - k_s^m - k_i^m) L^m = \left(\frac{2\pi n_p^m}{\lambda_p} - \frac{2\pi n_s^m}{\lambda_s} \cos \theta_s - \frac{2\pi n_i^m}{\lambda_i} \cos \theta_i \right) L^m \end{cases} \quad (3.1)$$

where δ and δ^m are the phases acquired inside the crystal and in the gap, respectively (see Equation (2.13)), k_j^m are SPDC wavevectors in the gap, n_j^m are the refractive indices of the medium in the gap ($j = s, i, p$), $\theta_{s,i}$ are the angles of the generated SPDC photons, $|\tau_i|$ is the amplitude transmission function of the medium at wavelength of the idler photons.

The phase of the interference pattern depends on the acquired phases of all three photons - signal, idler and pump, since they all travel through the medium in the gap.

Let us consider as a medium of interest a gas, which is transparent at the signal and pump wavelengths. Assuming no scattering and reflection losses, the visibility of the interference fringes is defined as $V = |\tau_i| = e^{-\alpha_i^m L^m / 2}$ (see Equations (2.27) and (2.34)).

Therefore, detecting the interference pattern of the signal SPDC photons in the visible range allows inferring the refractive index n_i^m and the absorption coefficient α_i^m

of the medium in the gap for the idler photons in IR range without resorting to IR optics and light sources.

3.1.1 Method

Firstly, the interference pattern is observed in a system with a vacuum gap (or air gap), where the refractive indices $n_{s,i,p}^{vac} = 1$ and the transmission coefficient $|\tau_i^{vac}| = 1$ are known. Under such conditions the interference pattern can be used as a reference, where, according to Equation (3.1), the interference pattern is determined as:

$$\begin{cases} P_s \propto \frac{1}{2} \left(\text{sinc}\left(\frac{\delta}{2}\right) \right)^2 \left(1 + 1 \cdot \cos\left(\delta + \delta^{vac}\right) \right) \\ \delta^{vac} = 2\pi \left(\frac{1}{\lambda_p} - \frac{1}{\lambda_s} \cos\theta_s - \frac{1}{\lambda_i} \cos\theta_i \right) L_m \end{cases} \quad (3.2)$$

When the medium under study is introduced in the interferometer, the interference pattern is modified:

$$\begin{cases} P_s^m \propto \frac{1}{2} \left(\text{sinc}\left(\frac{\delta}{2}\right) \right)^2 \left(1 + |\tau_i^m| \cdot \cos\left(\delta + \delta^m\right) \right) \\ \delta^m = 2\pi \left(\frac{n_p^m}{\lambda_p} - \frac{n_s^m}{\lambda_s} \cos\theta_s - \frac{n_i^m}{\lambda_i} \cos\theta_i \right) L_m \end{cases} \quad (3.3)$$

As a result, there is a phase shift of the interference pattern and the decrease of the visibility.

Furthermore, we consider small scattering angles θ_s and θ_i , which allows simplifying Equations (3.2) and (3.3) by expanding the sine and cosine functions as a series of small angles [78]:

$$\begin{aligned}
\Delta k(\omega_s, \theta_s) &= k_p - k_s \sqrt{1 - \left(\frac{\sin \theta_s}{n_s}\right)^2} - k_i \sqrt{1 - \left(\frac{k_s \sin \theta_s}{k_i n_s}\right)^2} = \\
&= k_p - k_s \left(1 - \frac{\theta_s^2}{2n_s^2} + \dots\right) - k_i \left(1 - \frac{1}{2} \left(\frac{k_s \theta_s}{k_i n_s}\right)^2 + \dots\right) = \\
&= \underbrace{k_p - k_s - k_i}_{=\vartheta_0} + \left(\frac{k_s \theta_s^2}{2n_s^2} + \frac{k_s^2 \theta_s^2}{2k_i n_s^2} + \dots\right) = \vartheta_0 + \frac{k_s \theta_s^2}{2n_s^2} \left(1 + \frac{k_s}{k_i}\right) = \\
&= \vartheta_0 + \frac{\pi \theta_s^2}{\lambda_s n_s} \left(1 + \frac{n_s \lambda_i}{n_i \lambda_s}\right) = \vartheta_0 + \frac{\pi \theta_s^2}{\lambda_s} \left(\frac{1}{n_s} + \frac{\lambda_i}{n_i \lambda_s}\right),
\end{aligned} \tag{3.4}$$

where we have neglected the higher order terms.

Note, that θ_i is rewritten in terms of θ_s according to the Equation (2.11).

The same is true for Δk_m at the gap. Therefore, we can define the following expressions from Equation (3.4):

$$\begin{aligned}
\Delta k(\omega_s, \theta_s) L/2 &= \left(\vartheta_0 + \frac{\pi \theta_s^2}{\lambda_s} \left(\frac{1}{n_s} + \frac{\lambda_i}{n_i \lambda_s}\right)\right) L/2 = \alpha_0 - C_1 \theta_s^2, \\
\Delta k(\omega_s, \theta_s)^m L^m &= \left(\vartheta_0^m + \frac{\pi \theta_s^2}{\lambda_s} \left(\frac{1}{n_s^m} + \frac{\lambda_i}{n_i^m \lambda_s}\right)\right) L^m = \beta_0 - C_2 \theta_s^2.
\end{aligned} \tag{3.5}$$

Using Equation (3.2), the measured interference pattern for a given wavelength is determined as:

$$P_s(\theta_s) = y_0 + C \text{sinc} \left(\alpha_0 - C_1 \theta_s^2 \right)^2 \left(1 + 1 \cdot \cos \left(\beta_0 - C_2 \theta_s^2 \right) \right), \tag{3.6}$$

where y_0 is the experimental background, C is the normalization factor, α_0, β_0 are additional phases, which do not depend on the angle θ_s , and $C_{1,2}$ are proportionality factors for the angular dependence. As the interference pattern with the vacuum gap is taken as a reference, the visibility is kept at $V_{ref} = 1$ (see Equation (3.2)), and the parameters $y_0, C, C_{1,2}, \alpha_0, \beta_0$ can be defined.

When the medium under study is introduced in the gap, the interference pattern in Equation (3.3) is given by:

$$P_s^m(\theta_s) = y_0 + C \operatorname{sinc} \left(\alpha_0 - C_1 \theta_s^2 \right)^2 \left(1 + V \cdot \cos \left(\beta_0 - C_2^m \theta_s^2 \right) \right). \quad (3.7)$$

Taking into account that the phase matching condition Δk does not change with the introduction of the medium, and that it is transparent for signal and pump photons, the parameters $y_0, C, C_1, \alpha_0, \beta_0$ are the same as in Equation (3.6). However, the parameters V, C_2^m are changed. The visibility of the interference pattern V depends on the transmission coefficient $|\tau_i^m|$ of the medium, and the phase shift of the interference pattern $(C_2 - C_2^m)$ depends on the refractive index n_i^m .

3.1.2 Experimental setup

Our experimental realization of the idea is shown in Figure 3.1. A continuous wave (CW) laser with $\lambda_p = 532$ nm wavelength (Spectra-Physics, Millennia V) with 100 mW power and with a beam waist $d = 2$ mm is used to pump the nonlinear crystals. Two Lithium Niobate crystals doped with 5% of Mg (MgO:LiNbO₃) with $L^m = 0.5$ mm thickness are used in the experiment. The crystals are placed parallel to each other and spaced approximately $L^m = 30$ mm apart.

In a type-I phase matching condition ($e \rightarrow o + o$) SPDC photon pairs may be generated in each crystal. Both crystals are put into a vacuum chamber, which has two optical windows. First we pump down the vacuum chamber to low pressures (about 20 mTorr) in order to avoid uncertainties such as unknown air composition. Then, we introduce CO₂ gas (purity $\geq 99.99\%$, National Oxygen) into the chamber at different pressures, which is monitored using a pressure gauge (Granville-Phillips, 275 Mini-Convectron).

We choose CO_2 gas, because it is widely used in environmental, biomedical and sensing applications [95, 96]. CO_2 gas is transparent at the visible wavelengths, but it absorbs light at mid-IR wavelengths, where we demonstrate the eligibility of the technique.

SPDC photon wavelengths are within the transparency range of the crystal, which is from $0.45 \mu\text{m}$ to $5 \mu\text{m}$. Wavelengths of the SPDC photons can be tuned depending on the crystal orientation (the angle θ_p between the pump wavevector and crystal optical axis). In our experiment we place the crystals in such way that the spectrum of IR photons of the SPDC coincides with the CO_2 absorption line at $4.27 \mu\text{m}$ (the orientation of the crystal corresponds to $\theta_p = 50.4^\circ$).

Fig. 3.1 Experimental setup using a nonlinear degenerate Mach-Zehnder interferometer with spectral selection. A CW laser at 532 nm wavelength is used to pump two Lithium Niobate (LiNbO_3) crystals, where SPDC field is generated. The crystals are put into a vacuum chamber with two optical windows. The CO_2 gas is injected into the vacuum chamber, and the pressure inside the vacuum chamber is monitored by the pressure gauge. Then, the pump is filtered out by the notch filters (indicated as pump filter here). The interference pattern is detected using spectrograph and CCD camera. The inset shows the propagation of the interfering SPDC wavevectors. As θ_s is small, down-converted photons from the first crystal stay within the beam diameter d , when they reach the second crystal. This condition provides the path indistinguishability between SPDC, which are occurred in two crystals.

At the detection system, the pump beam is filtered out using two notch filters (NF03-532E-25, Semrock), and the signal photons are focused onto a spectrograph slit (Acton, SpectraPro 2300i) with a $f = 500$ mm lens. As a result, the two-dimensional (wavelength-angular) interference pattern of SPDC photons is obtained by a CCD camera (Andor iXon3 897, the pixel area is $16 \times 16 \mu\text{m}^2$).

The wavelength resolution of the method is determined by the diffraction grating of the spectrograph, which in our case is 600 lines per mm, and by the CCD camera pixel size. In our setup each pixel in the x direction (wavelength axis) corresponds to $\delta\lambda_s = 0.08$ nm. The wavelength resolution at the IR range also depends on the phase matching condition (or observed signal photon wavelengths λ_s):

$$\Delta\lambda_i = \frac{\Delta\lambda_s\lambda_p}{(\lambda_s - \lambda_p)} - \frac{\Delta\lambda_s\lambda_s\lambda_p}{(\lambda_s - \lambda_p)^2} = -\frac{\Delta\lambda_s\lambda_p^2}{(\lambda_s - \lambda_p)^2}, \quad (3.8)$$

where $\lambda_p = 532$ nm is pump wavelength. Therefore, the wavelength resolution at the IR region is $\Delta\lambda_i = 3.8$ nm (or 2.2 cm^{-1}) for $\theta_p = 50.4^\circ$ phase matching angle ($\lambda_s = 609$ nm, $\lambda_i = 4208.6$ nm).

Using a $f = 500$ mm lens before the CCD camera allows us to measure the SPDC angles up to 1° . A longer focal length leads to a larger image on the CCD camera. Thus, the angular resolution is defined by the focal length f of the lens and the CCD camera pixel size. In our experiment each pixel in the y direction (angular axis) corresponds to $\delta\theta_s = 0.002^\circ$.

By increasing the gap length between crystals, the accuracy in determining the refractive index and absorption coefficient can be improved. Indeed, increasing the gap length L^m decreases the periodicity of the interference fringes. Thereby, phase shift at the gap δ^m can be detected more precisely. Increasing the base of the interferometer also increases the absorption losses, which improves the sensitivity especially at low

concentrations of the gas. However, in this case, the problem of the resolving of the interference fringes appears.

3.1.3 Measured data

We study the absorption line of CO₂ gas at a wavelength of $\lambda_i = 4.27 \mu\text{m}$. First, we perform measurements at the range of wavelengths where idler photons of the SPDC do not coincide with the absorption lines of the CO₂ gas. In this case the crystal is oriented at $\theta_p = 51.8^\circ$, where the signal and idler photons are at $\lambda_s = 620 \text{ nm}$ and $\lambda_i = 3748 \text{ nm}$, respectively (see Equation (2.1)).

The wavelength-angular interference patterns of the signal SPDC photons are shown in Figure 3.2. Figure 3.2 (a) corresponds to the data with the vacuum gap between crystals, Figure 3.2 (b) shows the interference pattern with injected CO₂ gas at 30 Torr pressure.

Figure 3.3 shows the vertical cross-sections of the two-dimensional interference patterns in Figure 3.2 at $\lambda_s = 620.8 \text{ nm}$ wavelength. The black curve corresponds to data with a vacuum gap, the red curve shows the data with CO₂ gas.

Introducing the CO₂ gas changes the refractive index n_j^m in the gap, thus shifting the interference fringes. However, the visibility of the interference pattern remains the same, as idler photons are within the transparency range of the CO₂ gas.

Next, we set the the idler wavelength to coincide with the absorption line of the CO₂ gas at $\lambda_i = 4.27 \mu\text{m}$. The wavelength of the signal photons is $\lambda_s = 607.7 \text{ nm}$ (see Equation (2.1)). The phase matching angle is $\theta_p = 50.4^\circ$. The interference patterns for the signal SPDC photons are shown in Figure 3.4.

The wavelength $\lambda_i = 4236.5 \text{ nm}$ ($\lambda_s = 608.4 \text{ nm}$) is close to the center of the absorption line of the CO₂ gas (see Figure 3.4 (c)). As a result, there is the small change in refractive index resulting in a small phase shift of the interference fringes.

Fig. 3.2 **Two-dimensional wavelength-angular interference pattern of the signal SPDC photons, when the conjugate wavelength of idler photons does not coincide with the absorption line of the CO₂ gas.** (a) Interference pattern with the vacuum gap between crystals. (b) Interference pattern with injected CO₂ gas having 30 Torr pressure. Both graphs are plotted using the same scale, which is shown at the right-hand side from the graph (b).

Fig. 3.3 **Cross-sections of the two-dimensional wavelength-angular interference pattern of the signal SPDC photons at $\lambda_s = 620.8$ nm wavelength.** The black and red curves show data with the vacuum gap and the gap filled with CO₂ gas, respectively.

However, the visibility of the interference pattern is significantly reduced due to the absorption of CO_2 gas at the wavelength of idler photons.

The wavelength $\lambda_i = 4160$ ($\lambda_s = 610$ nm) nm is detuned from the center of the absorption line (see Figure 3.4 (d)). Therefore, there is a clear shift of the interference fringes, but the visibility is not significantly reduced.

Fig. 3.4 Two-dimensional wavelength-angular interference pattern of the signal SPDC photons, when the conjugate wavelength of idler photons coincides with the absorption line of the CO_2 gas. (a) Interference pattern with a vacuum gap between crystals. (b) Interference pattern with injected CO_2 gas at 10.5 Torr pressure. (c) Cross-section of two-dimensional interference patterns at the $\lambda_s = 608.4$ nm wavelength ($\lambda_i = 4236.5$ nm). (d) Cross-sections of two-dimensional interference patterns at the $\lambda_s = 610$ nm wavelength ($\lambda_i = 4160.5$ nm). In (c) and (d) black and red curves correspond to the data with the vacuum gap and the gap filled with CO_2 gas, respectively.

3.1.4 Results and Discussion

First, we obtain the cross-sections of the interference pattern at different wavelengths of signal photons similar to those shown in Figures 3.3 and 3.4 (c) and (d). We average the data within ten columns of pixels, corresponding to $\Delta\lambda_s = 0.8$ nm ($\Delta\lambda_i = 38$ nm or 22 cm⁻¹), to get higher signal-to-noise ratio. Then, we fit the data using Equations (3.6) and (3.7). The fitting of the data at $\lambda_s = 620$ nm, 610 nm and 608.4 nm are shown in Figures 3.5, 3.6 and 3.7, respectively (with $R^2 = 95\%$). Fitting was made in Origin software using the built-in method of least means squares.

The properties of CO₂ gas at the visible wavelength region ($n_{p,s}$) are known [97, 98]. Then, it is possible to calculate the properties of the medium for the IR region (n_i^m and $|\tau_i^m|$).

We start by considering the data with the vacuum gap. We need to find the phase matching angle θ_p and gap length L^m for further calculations. The first set of data corresponds to $\theta_p = 51.8^\circ$ and $L^m = 31$ mm, the second set corresponds to $\theta_p = 50.4^\circ$ and $L^m = 30$ mm.

Fig. 3.5 **Cross-section of the wavelength-angular interference pattern at $\lambda_s = 620$ nm.** (a) with the vacuum gap and (b) with the gap filled with CO₂ gas at 30 Torr pressure. The black curve correspond to the experimental data, the red dashed line shows the fitting curve.

Fig. 3.6 **Cross-section of the wavelength-angular interference pattern at $\lambda_s = 610 \text{ nm}$. (a) with the vacuum gap and (b) with the gap filled with CO_2 gas at 10.5 Torr pressure. The black curve correspond to the experimental data, the red dashed line shows the fitting curve.**

Fig. 3.7 **Cross-section of the wavelength-angular interference pattern at $\lambda_s = 608.4 \text{ nm}$. (a) with the vacuum gap and (b) with the gap filled with CO_2 gas at 10.5 Torr pressure. The black curve correspond to the experimental data, the red dashed line shows the fitting curve.**

Then, the refractive index and absorption coefficient at the wavelength of idler photons are calculated. Here, we consider a linear dependence of the refractive indices at the visible range on pressure:

$$n(P) = 1 + \frac{P(n_0 - 1)}{P_0 \left(1 + \frac{T - T_0}{T_0}\right)}, \quad (3.9)$$

where T is the temperature of the gas (in our case it is at room temperature $T = 300$ K), $T_0 = 273$ K, and n_0 is the refractive index of the CO₂ gas at atmospheric pressure P_0 .

Note that the need for prior knowledge of refractive indices at the visible range $n_{s,p}^m$ can be overcome by splitting the signal and idler photons after the first crystal and then recombining them at the second crystal. A medium can be inserted only into the path of the idler photons. This configuration of the interferometer is described in later Chapters.

The value of the absorption coefficient α_i^m is calculated from the visibility of the interference pattern, taking into account the reference visibility which is determined from measurements with the vacuum gap as discussed in section 3.1.1.

Experimental results with CO₂ gas at the pressure of 10.5 Torr are shown in Figure 3.8 (a). The absorption line α_i^m has a peak at 4.27 μm with a full-width at half-maximum (FWHM) of 140 nm. The experimental data are in good agreement with the theoretical value taken from HITRAN database [99].

Figure 3.8 (b) shows the dependence of the refractive index of the CO₂ gas on the wavelength. The refractive index was calculated from the relative shift of the interference fringes. The experimental points are in good agreement with the theory based on the Kramers–Kronig equations and absorption coefficient data [100].

Figure 3.9 shows the absorption coefficient α_i^m and refractive index n_i^m as a function of the gas pressure at different idler wavelengths λ_i . These quantities exhibit nonlinear

Fig. 3.8 (a) Refractive index and (b) absorption coefficient of the CO₂ gas at 10.5 Torr pressure depending on wavelength. The blue points correspond to the experimental data. The red line in (a) shows the theoretical value of the absorption coefficient. The red curve in (b) shows the refractive index calculated using Kramers-Kronig relations and database values for the absorption coefficient.

Fig. 3.9 (a) Refractive index and (b) absorption coefficient depending on CO₂ gas pressure. The different colors correspond to different wavelengths of the idler photons λ_i . In Figure (a) wavelengths are shown in terms of the detuning from the center of the absorption line at $\lambda_0 = 4.27 \mu\text{m}$.

variations in the vicinity of the absorption line, which can be explained by different mechanisms of line broadening at different pressures [101, 102]. There is a Doppler line broadening due to the motion of molecules at lower pressures, and the line has a Lorentzian shape due to collision broadening at higher pressures. But far from the absorption line the measurement of the refractive index shows a linear dependence on pressure.

In the current experimental setup the calculation accuracy for the absorption coefficient varies in the range $\Delta\alpha_i^m = 0.02 - 0.2 \text{ cm}^{-1}$ depending on the absorption value. The refractive index measurement accuracy is $\Delta n_i^m = 5 \cdot 10^{-6}$.

3.2 Increasing the signal-to-noise ratio of the method

3.2.1 Method

The method described in the previous section uses cost-efficient and well developed visible light components. However, the technique uses spectral and spatial filtering of the signal, which is performed by a spectrograph and a slit. The filtering cuts down the signal throughput and reduces the signal-to-noise ratio of the method.

In this section, we demonstrate an approach to increase the signal-to-noise ratio of the method by eliminating the need for the spectrograph and the slit. This approach reduces the system footprint and makes it more affordable.

The scheme of the interferometer is shown in Figure 2.7 (b). In this case the spectral filtering is eliminated, and the interference pattern is determined by the integration of Equation (3.1) over the frequency range $d\omega$:

$$P_s(\theta_s) \propto \int d\omega \left(\text{sinc}\left(\frac{\delta}{2}\right) \right)^2 \left(1 + |\tau_i^m| \cos(\delta + \delta^m + \pi) \right), \quad (3.10)$$

where additional phase π comes from the reflection from the mirror. Here, the periodicity of the interference pattern depends on the phase under the cosine function. The visibility depends on both the phase under cosine δ_m and on the amplitude transmission coefficient τ_i^m .

When the slit is eliminated, the whole two-dimensional SPDC spectrum is detected by CCD camera. The SPDC spectrum has circularly symmetric shape and it is modulated due to the nonlinear interference. Capturing the whole spectrum allows us to measure interference fringes across all the radial directions at once, which improves the signal-to-noise ratio of the method.

The interference fringes can be analyzed using the following function:

$$P_s(\theta_s) = y_0 + A \int d\omega \left(\text{sinc}\left(\frac{\delta}{2}\right) \right)^2 \left(1 + |\tau_i^m| V(\theta_s) \cos(\delta + \delta^m + \pi) \right), \quad (3.11)$$

where y_0 is the background level, and A is the magnitude of the interference fringes, $V(\theta_s)$ is an additional visibility function, which takes into account the decrease of the visibility with the angle due to the spatial overlap of the beams (as in Equation (2.33)) [69].

This integral cannot be solved analytically, and has to be simulated numerically. In this method, the interference visibility depends on many parameters besides properties of the medium between crystals. The gap length L^m , the phase matching angle θ_p and the crystal thickness L also determine the form of the interference pattern. All these parameters have to be taken into account at the analysis.

The case with the vacuum gap is a convenient reference point with known refractive indices $n_{p,s,i}^{vac} = 1$ and amplitude transmission coefficient $|\tau_i^{vac}| = 1$. Measurements with the vacuum allow to account for optical losses for the signal, idler and pump photons in the nonlinear crystals. By fitting the experimental results with Equation (3.11) we determine parameters δ , L^m , y_0 , A and $V(\theta_s)$.

The parameters $|\tau_i^m|$ and δ^m change after introducing a medium into the gap, while all other parameters remain the same. Taking n_p^m and n_s^m from the literature, both n_i^m and α_i^m can be inferred from the fitting procedure.

3.2.2 Experimental setup

The experimental setup of the method is shown in Figure 3.10. Similarly as in the previous experiment, we use a continuous wave (CW) laser ($\lambda_s = 532$ nm, 30 mW power). However here, instead of two nonlinear crystals, we use one crystal (MgO:LiNbO₃, $L = 1$ mm, for type-I SPDC with the cut angle at $\theta_c = 50^\circ$) and a silver mirror. The SPDC field is generated at the first pass of the pump beam through the crystal. Then, pump beam and SPDC field are reflected back by the mirror, acquiring an additional phase shift π . The SPDC field also occurs during the second pass of the pump through the crystal.

The crystal is placed on a rotation stage (ThorLabs, with 0.2° step accuracy). The wavelength tunability of the method is achieved by tilting the crystal. Similarly as in previous experiment, the interferometer is put into a vacuum chamber. But here, the interference pattern is directly focused in to the CCD camera (Andor, iXon3 897, same as before) by a lens F with $f = 300$ mm without using the spectrograph and a slit. A bandpass filter BP (605 ± 15 nm; Semrock) is filtering the noise from the stray light.

The technique is again tested with an absorption line of the CO₂ gas. We focus our study on a strong absorption line of the CO₂ at the wavelength of $\lambda_i = 4.27$ μm (the conjugate signal wavelength is about $\lambda_s = 607.7$ nm). The absorption coefficient and the refractive index are measured in the vicinity of this line.

We obtain the interference pattern with CCD camera with 100 seconds acquisition time. The background noise is measured by tilting the crystal so that the SPDC signal does not pass through the bandpass filter.

The SPDC photons are filtered by a dichroic mirror, a notch filter, a vertically oriented glan-prism polarizer and the bandpass filter BP. Then interference pattern is directly observed by the CCD camera.

Fig. 3.10 Experimental setup using nonlinear degenerate Mach-Zehnder interferometer without spectral selection. A CW laser (532 nm wavelength) is directed into the vacuum chamber using the dichroic mirror DM. The laser is prepared having horizontal (H) polarization. A nonlinear crystal and a mirror are placed inside the vacuum chamber. SPDC field is generated at the forward and backward passes of the pump through the nonlinear crystal.

3.2.3 Results and Discussion

Experimentally obtained interference patterns with and without CO₂ gas are presented in Figure 3.11. Without the slit and the spectrograph, we observe the full two-dimensional picture of the interference pattern. Measurements are done for $\theta_p = 50.5^\circ$ phase matching angle, which yields $\lambda_s = 608.4 - 611.3$ nm and $\lambda_i = 4100 - 4240$ nm, where the CO₂ gas has a strong change in the refractive index, but comparatively low absorption. Therefore, interference fringes experience a significant phase shift.

First, the data processing is performed to eliminate the artifacts and reduce the speckle noise using a program written in Python. Then, the averaging of the signal along the azimuthal angles is performed. The data is averaged across the whole spectrum of the SPDC (about 70 points per angle at $\theta_s = 0.5^\circ$), which leads to 8 times increase in the signal-to-noise ratio compared to the case, when the slit is used.

The experimental parameters δ , L^m , y_0 , A and $V(\theta_s)$ are found using Equation (3.11) by fitting the data for the interference fringes obtained with the vacuum gap. The fitting is done using a program written in Python. The program finds the best fitting of the experimental data using the least mean square algorithm (see Appendix C). Next, we analyze the data obtained with the CO_2 gas. At this step the parameters δ , L^m , y_0 , A and $V(\theta_s)$ are fixed and the parameters δ^m and $|\tau_i^m|$ are unknown. First, we evaluate δ^m , which includes only one unknown parameter - the refractive index at the

Fig. 3.11 Experimental interference patterns of SPDC photons obtained in the interferometer with one crystal and a mirror with $L^m/2 = 10$ mm and $\theta_p = 50.5$, where signal and idler waves have $\lambda_s = 608.4 - 611.3$ nm and $\lambda_i = 4100 - 4240$ nm wavelengths, respectively. The interference pattern (a) with the vacuum gap; (b) with the gap with CO_2 gas at 7 Torr pressure. The red arrow in (a) indicates the averaging over azimuthal angles. (c) Interference fringes for the data with the vacuum gap (d) and with the gap filled with CO_2 gas at 7 Torr pressure. The red bars indicate the standard deviation of the experimental data. The black dots show the experimental points, the red dashed lines show the fitted curves.

idler photon wavelengths n_i^m . Since the width of the CO₂ absorption line is comparable with the SPDC linewidth, we take into account that the refractive index has frequency dependence. We take the refractive index according to the Lorentz model [103]. We also use partial refractive indices $n_{(p,s,i0)}^m$ to account for its pressure dependence:

$$n_{(p,s,i0)}^m = \left(1 - \frac{P_{CO2}}{P_{atm}}\right)n_{vac} + \frac{P_{CO2}}{P_{atm}}n_{CO2(p,s,i)}, \quad (3.12)$$

where P_{atm} is the atmosphere pressure, P_{CO2} is the CO₂ pressure, $n_{CO2(p,s,i)}$ is the CO₂ refractive index at the atmospheric pressure, which are taken from literature [97, 98]. Then, we fit the data by Equation (3.11).

Figures 3.11 (c) and (d), present the experimental data and the numerical simulations for the gap without and with the CO₂ gas, respectively. The experimental points show a good agreement with the theoretical simulations.

Figure 3.12 shows the experimentally measured n_i^m and α_i^m of the CO₂ gas. Figure 3.12 also presents the theoretical curve, which is calculated using the Kramers-Kronig relations and the absorption coefficients given by HITRAN database [99]. Our method has the spectral resolution of 80 cm⁻¹, which is determined by the natural linewidth of the SPDC spectrum. Therefore, we take the theory for the absorption coefficient from the database with the following parameters: the calculation step is 0.01 cm⁻¹, the rectangular function resolution is 80 cm⁻¹.

The dependences of the refractive index and the absorption coefficient on the CO₂ pressure is shown in Figure 3.13. Theory is taken from the HITRAN database with the same input parameters. The measured refractive indices and the absorption coefficients are in good agreement with the theory.

In the previous experiment, the resolution was 2.2 cm⁻¹, which was determined by the resolution of the spectrometer. The current method has the spectral resolution of 80 cm⁻¹, which is defined by the natural linewidth of the SPDC. However, a longer

Fig. 3.12 Measured values for (a) the refractive index and (b) the absorption coefficient of the CO_2 gas at 7 Torr pressure. The black dots represent calculated values, the red lines indicate theoretical curves. The top horizontal axis indicates corresponding wavelengths of the measured signal photons, and the bottom horizontal axis shows the actual wavelength at the IR (for idler photons).

Fig. 3.13 Pressure dependence of (a) the refractive index and (b) the absorption coefficient at different wavelengths. The legend shows the detuning wavelength from $\lambda_0 = 4266$ nm. The theory for the absorption coefficient is shown by solid lines. Pressure dependence of (a) the refractive index and (b) the absorption coefficient at different wavelengths, which are shown as detuning wavelengths from λ_0 . Because of the small value of the absorption coefficient, the data for 220 nm and - 222 nm detuning wavelengths almost coincide.

nonlinear crystal can be used for narrowing the SPDC spectrum. For comparison, the crystal with $L = 5$ mm length would give a resolution of 15 cm^{-1} . Using a longer

SPDC crystals also increases the SPDC signal, which scales quadratically with the crystal length (see Equation (2.15)).

The experimental data and the theory of the refractive index and the absorption coefficient in a good agreement with each other. Some differences arise due to the low sensitivity of our method at pressures below 5 Torr.

3.3 Summary

In this chapter we demonstrated an IR spectroscopy technique based on the nonlinear interference of frequency correlated photons produced via SPDC. It overcomes the limitations of conventional IR techniques, and requires only a simple setup with accessible visible-range detectors and lasers. This approach allows direct measurement of the refractive index and the absorption coefficient of a medium in the broadband IR range by detecting photons in the visible range. The tunability of the SPDC wavelength allows the tailoring of a measurement range to a specific application, including far-infrared and terahertz (THz) ranges [104, 105]. This approach represents a practical alternative to conventional IR techniques.

At the first part of the chapter we obtain a high measurement accuracies of the refractive index ($\Delta n_i^m = 5 \cdot 10^{-6}$) and the absorption coefficient ($\Delta \alpha_i^m = 0.02 - 0.2 \text{ cm}^{-1}$), which are on par with conventional IR methods. The spectral resolution of the method is 2.2 cm^{-1} (at 2330 cm^{-1}).

At the second part of the chapter we propose an approach to increase the signal-to-noise ratio of the nonlinear interference method via parallel readout of multiple spatial and frequency modes. The main advantage of this approach in comparison with the previous scheme is that it excludes the need for the spatial and spectral selection. This allows one to obtain the whole picture of the interference pattern and makes the setup economical and compact. In the current scheme the refractive index is measured with

an accuracy of $\Delta n_i^m = 5 \cdot 10^{-6}$, and the absorption coefficient with an accuracy of $\Delta \alpha_i^m = 0.05 \text{ cm}^{-1}$. The spectral resolution of the method is 80 cm^{-1} (at 2330 cm^{-1}), which can be improved by using longer crystals.

Chapter 4

Measurement of infrared optical constants using a nonlinear Michelson interferometer

The nonlinear spectroscopy schemes described in Chapter 3 consisted of two nonlinear crystals, which were set sequentially in a common pump beam. A sample was placed between the crystals and all the three photons (pump, signal, and idler) passed through the sample. The absorption spectrum of the sample in the IR was inferred from the interference pattern of the visible photons.

The described methods do not require the use of IR equipment, and are highly stable due to the degenerate arms of the interferometer. However, it has several practical limitations. The major issue is that the sample has to be transparent at wavelengths of signal and pump photons, and its optical properties at these wavelengths have to be known in advance. There is also a risk that the pump laser damages the sample, and light scattering in the sample may limit the performance of spectroscopic and interferometric instruments.

Fig. 4.1 **Scheme of the nonlinear Michelson interferometer.** The pump beam generates SPDC field at the first passage through the nonlinear crystal ($s1$, $i1$). SPDC field is split by the dichroic mirror into different arms. Pump beam and SPDC field are recombined at the crystal by the mirrors M_s and M_i . Another SPDC field ($s2$, $i2$) is generated by the pump at the second passage through the nonlinear crystal. The sample is inserted into the path of idler photons $i1$.

In this chapter, we introduce an alternative approach to IR spectroscopy with visible light, which is free of the limitations mentioned above. We build a setup in the form of a nonlinear Michelson interferometer, where the signal (in the visible range) and the idler (in the IR range) photons are generated collinearly, split using a dichroic beam splitter and then reflected back into the crystal by two mirrors (see Figure 4.1). The medium of interest, placed in the path of the idler photons, modifies the interference pattern for the signal photons. We infer the transmission coefficient of the sample and its refractive index in the IR range from the modified interference pattern of visible photons. Although the alignment of this scheme is more challenging in comparison to the degenerate Mach-Zehnder interferometer, it is appealing from a practical standpoint. Unique benefits of this nonlinear Michelson interferometer compared to the ones reported in Chapter 3 are:

1. It works for samples opaque in the visible range;
2. It does not require a prior knowledge of the sample properties in the visible range;
3. The pump beam will not damage the sample;

4. The pump and the signal photons are not scattered by the sample. Thus, in experiments with highly scattering samples, a better sensitivity and larger penetration depth can be achieved by probing the samples only by IR light.

The difference between this scheme and the one described in section 2.3.3 is that here we use a collinear geometry of the interferometer, which is more flexible in alignment, since the pump beam can be used as a reference for the alignment procedure. This work further adds to the number of optical characterization techniques, which benefit from quantum enhancement effects [53, 54, 106–109].

4.1 Method

Let us consider the scheme shown in Figure 4.1, which represents a nonlinear version of the Michelson interferometer. At the first pass through the nonlinear crystal, the pump creates the SPDC field. Next, the SPDC field is split using a dichroic mirror and reflected back to the crystal by the mirrors M_s and M_i . Pump and signal beams propagate together. The reflected pump beam generates the SPDC field also at the second pass through the crystal. A medium of interest is inserted into the path of idler photons.

Similar to section 2.2, the state vector of the SPDC field in the interferometer is given by a superposition of state vectors of the SPDC generated from the first "1" and the second "2" pass of the pump through the nonlinear crystal:

$$\begin{aligned}
 |\Psi\rangle = |\psi_1\rangle + |\psi_2\rangle = |vac\rangle + \eta \sum_{\mathbf{k}_s, \mathbf{k}_i} F(\mathbf{k}_s, \mathbf{k}_i) a_{\mathbf{k}_{s1}}^+ a_{\mathbf{k}_{i1}}^+ |vac\rangle + \\
 + \eta e^{i\phi_p} \sum_{\mathbf{k}_s, \mathbf{k}_i} F(\mathbf{k}_s, \mathbf{k}_i) a_{\mathbf{k}_{s2}}^+ a_{\mathbf{k}_{i2}}^+ |vac\rangle,
 \end{aligned} \tag{4.1}$$

Then, following same steps as described in section 2.2 (see Equation 2.26), the interference pattern is given by:

$$P_s(\Delta t) \propto 2 \left(1 + |\tau_i^m|^2 |\mu(\Delta t)| \cos \left(\phi_p - \phi_s - \phi_i + \arg(\tau_i^{m2}) + \arg(\mu(\Delta t)) \right) \right), \quad (4.3)$$

$$\mu(\Delta t) = \int |F(\Omega)|^2 \exp(-i\Omega\Delta t) d\Omega,$$

where $\mu(\Delta t)$ is the normalized correlation function of the SPDC [24], and Δt is the time delay between signal $s1$ and idler $i1$ photons in the interferometer. The illustration of the interference pattern is shown in Figure 2.5.

The delay Δt is equal to zero when the optical path lengths of signal and idler photons in the interferometer are balanced (see Equation (2.31)):

$$n_s L + n_s^m L_s^m = n_i L + n_i^m L_s^m, \quad (4.4)$$

where $n_{s,i}$ are the refractive indices of the nonlinear crystal at signal and idler wavelengths, $n_{s,i}^m$ are the refractive indices of the medium at signal and idler wavelengths, and $L_{s,i}^m$ are the path lengths for signal and idler photons in the interferometer.

If the accumulated phases of the signal and of the pump photons are fixed, then the variation of the photon count rate $P_s(\Delta t)$ is determined by the phase of the idler photons $\phi_i = 2\frac{2\pi n_i^m}{\lambda_i} \Delta z$, where Δz is the displacement of the IR mirror M_i . Then the visibility of the interference fringes is given by:

$$V = \frac{P_s^{max} - P_s^{min}}{P_s^{max} + P_s^{min}} = |\tau_i^m|^2 |\mu(\Delta t)|. \quad (4.5)$$

From Equation (4.5) it follows that the transmission coefficient of the idler (IR) photons through the medium can be inferred from the interference visibility of signal photons detected in the visible range.

The experimental procedure for measuring the transmission spectra of the medium is as follows. First, we equalize the interferometer arms and measure the interference with the air as a reference medium $n_{s,i}^m = n_{s,i}^{air} = 1$. From the measurements of the interference visibility V_{ref} we infer the total intrinsic losses of our system $|\tau_i^{app}|^2$, see Equation (4.5). Then we insert the medium of interest and re-balance the interferometer arms to compensate for the extra delay:

$$\delta L^m = 2(n_i^m - n_i^{air})L^m, \quad (4.6)$$

where L^m is the thickness of the medium here. Then, we measure the interference visibility again and infer its relative change:

$$\frac{V}{V_{ref}} = \frac{|\tau_i^{app}|^2 |\tau_i^m|^2}{|\tau_i^{app}|^2} = |\tau_i^m|^2 \equiv T_i^m. \quad (4.7)$$

We infer the refractive index and the transmission coefficient of the medium in the IR range from the change of the optical path and the visibility, respectively (see Equations (4.6) and (4.7)). There are always reflection losses at the boundaries in addition to the absorption losses inside the medium. Therefore, from the obtained refractive index and transmission coefficient values, we can infer the reflection coefficient R_i^m and absorption α_i^m of the medium at the corresponding wavelengths:

$$\begin{aligned} |r_i^m|^2 \equiv R_i^m &= \left(\frac{n_i^m - n_i^{air}}{n_i^m + n_i^{air}} \right)^2, \\ \frac{V}{V_{ref}} = |\tau_i^m|^2 &= \left(\left(1 - |r_i^m|^2 \right) \exp(\alpha_i^m L^m / 2) \right)^2 = \left(1 - R_i^m \right)^2 \exp(\alpha_i^m L^m), \end{aligned} \quad (4.8)$$

where R_i^m is the reflection coefficient calculated at normal incidence.

4.2 Experimental setup

The full experimental setup is shown in Figure 4.3. A CW laser with $\lambda_p = 532$ nm wavelength is used as the pump (Laser Quantum Torus, power 150 mW, coherence length about 100 meters). A dichroic mirror DM_1 (Semrock) reflects the pump beam towards the nonlinear interferometer and transmits the signal photons in the range of 562 – 950 nm. The nonlinear Michelson interferometer consists of a $MgO:LiNbO_3$ crystal with $L = 0.5$ mm length (Castech), a gold-coated dichroic mirror DM_2 (ISP Optics), and two spherical mirrors ($f = 50$ mm, Thorlabs): one is silver coated M_s , and another one is gold coated M_i .

The pump beam is focused into the crystal by the lens F ($f = 200$ mm). The SPDC field is generated via type-I ($e \rightarrow o + o$) phase matching in a collinear, frequency non-degenerate regime. We tune the central wavelengths of the down-converted photons by tilting the crystal. The idler photon wavelength is inferred from the wavelength of signal photons according to Equation (2.1). The dichroic mirror DM_2 splits SPDC field into two arms. Mirrors M_s and M_i recombine the SPDC field and the pump beam back into the crystal. The pump beam creates SPDC field at the backward pass through the nonlinear crystal.

In the detection part, the signal photons are separated from the pump by the dichroic mirror DM_1 and the notch filter NF (Semrock). The signal photons are then collimated by a lens F_2 ($f = 200$ mm). The interference pattern can be detected in two ways:

1. Using a silicon CCD camera (Andor, iXon3 897). The CCD is located at the focal plane of the lens, and it images the beam of signal photons. Measurements with the CCD camera facilitate alignment of the setup.

2. By a single photon avalanche photodiode (APD, Perkin Elmer) preceded or not preceded by a home-built monochromator. The monochromator allows performing wavelength selective measurements at different wavelengths within the spectrum of signal SPDC photons.

The visibility of the interference pattern depends on the spatial overlap of the interfering photons [69]. The intrinsic divergence of the SPDC output results in different phases being acquired by transverse wavevector components, which leads to pronounced ring structure, as we observed in Chapter 3. To circumvent this problem, we focus the pump beam inside the crystal, so that the divergence of the down-converted photons is

Fig. 4.3 Experimental setup using the nonlinear Michelson interferometer for IR spectroscopy measurements. A CW laser at 532 nm wavelength is used as a pump for a LiNbO₃ crystal, where SPDC field is generated. The dichroic mirror DM₂ splits the SPDC field into two arms. Spherical mirrors M_s and M_i ($f = 50$ mm) reflect the beams back into the crystal. A sample is placed in the IR arm of the interferometer between DM₂ and M_i. Using the flipping mirror, we capture the interference pattern either by a CCD camera or by a single photon detector (APD) preceded by a monochromator. The signal is filtered using a notch filter NF and a bandpass filter BP.

defined by the divergence of the pump beam (see Equation (2.10)). We use spherical mirrors $M_{s,i}$, positioned at a distance of their curvature radius ($R = 2f$) from the crystal to form a one-to-one image of the pump waist in the crystal. The use of metallic spherical mirrors eliminates the problem of chromatic dispersion. In this way, SPDC beams produced in two passes of the pump photons through the nonlinear crystal can be perfectly overlapped.

The interference pattern obtained by the CCD camera is shown in Figure 4.4. The spatial modes of the SPDC beams are not efficiently overlapped, if plane mirrors are used in the interferometer (see Figure 4.4 (a)). In this case the interference is present only for limited angles of SPDC in accordance with Equation (2.33). When spherical mirrors are introduced into the interferometer arms, it is possible to efficiently overlap the spatial modes of the SPDC and increase the visibility of the interference pattern (see Figure 4.4 (b)). Figure 4.4 (c) shows the case when one of the mirrors $M_{s,i}$ reflect the SPDC photons not to the same path in the interferometer. In this case, the visibility decreases in comparison with Figure 4.4 (b).

Then, to measure the refractive index n_i^m , and transmission coefficient T_i^m of samples we perform measurements of the interference pattern by the APD. First, we bypass the monochromator and move the mirror M_i using a motorized translation

Fig. 4.4 Interference patterns obtained by the CCD camera, when there are plane mirrors (a) and spherical mirrors (b) in the interferometer arms, respectively; (c) when one of the spherical mirrors reflect the beam not to the same path in the interferometer.

stage to detect the envelopes of the interference pattern (as in Figure 2.5). From the shift of the maxima of the interference patterns with and without the sample, we infer the refractive index n_i^m in the IR range according to Equation (4.6).

Next, we perform a fine scan of the interference fringes with and without the sample. Here, the signal photons are spectrally filtered by the monochromator. The resolution of the monochromator for visible photons is about 0.25 nm (2.5 nm (5.2 cm⁻¹) at 2200 nm (4.55 cm⁻¹) in the IR), which is on par with commercially available FTIR systems. Therefore, the coherence time of the SPDC photons increases, significantly reducing the accuracy of the optical delay measurements introduced by the sample. Here, we translate the mirror M_i using a piezo actuator and measure visibility of the interference fringes. Then, we infer the transmission coefficient of the sample T_i^m at the wavelength of idler photons according to Equation (4.7).

We observe that the phase drift in the interferometer was slower than the typical measurement time of one fringe (~ 30 sec). It is certainly possible to implement active stabilization of the interferometer, for instance, by monitoring the intensity of the residual pump beam exiting from the empty port of the dichroic mirror DM₂, but it was not necessary in our measurements.

4.3 Results and discussion

We apply our technique to study four different samples: polydimethylsiloxane (PDMS), BK7 glass and pure silicon windows with and without anti-reflection (AR) coating (Edmund Optics). For a proof-of-concept demonstration of our technique we choose the spectral range within 2100 – 2900 nm, which corresponds to the observation of signal photons at the range of 710 – 655 nm.

First, we measure the envelope of the interference pattern without the sample. The dependence of the APD photocounts on the position of the mirror M_s is shown in

Figure 4.5 (black squares). Next, we insert a polymer sample (Polydimethylsiloxane (PDMS) with $143\ \mu\text{m}$ thickness) in the path of the IR beam. The interference pattern shifts approximately to $120\ \mu\text{m}$, and its visibility reduces (see Figure 4.5, red circles). From the change of the optical path length, we infer the refractive index of PDMS in the IR range (see Table 4.1).

The envelope of the interference pattern is given by the first-order correlation-function of the SPDC $|\mu(\Delta t)|$. In Figure 4.5 we plot the theoretical curve of the envelope of the interference pattern with parameters obtained from the experimentally measured SPDC spectrum (see Figure 4.6) and the visibility values. Since the SPDC spectrum has a sinc^2 form (see Equation (2.10)), the correlation function has a triangular

Fig. 4.5 Measured photocount rate of the APD versus the position of the mirror M_i (without the monochromator). Black squares show the data with the air, and red circles show the data obtained with a $143\ \mu\text{m}$ thick PDMS sample. The thick solid lines show the envelope function given by $|\mu(\Delta t)|$ in Equation (4.3). The envelope is plotted using measured experimental parameters of the interference visibility and the SPDC coherence length. The inset shows the interference fringes observed by fine translation of the mirror M_i using a piezo actuator (with the monochromator). The solid lines show the data fitted by cosine-functions according to the first Equation in (4.3) with $R^2 = 0.95$. The absorption coefficient is inferred from the amplitude of the fitting function.

shape, with amplitude given by the visibility of the interference pattern and with the width given by the coherence length of the SPDC photons. The spectrum of the SPDC signal photons shown in Figure 4.6, where the width of the spectrum at $\lambda_s = 694.4 \pm 0.1$ nm is $\Delta\lambda_s = 5.8 \pm 0.2$ nm, which corresponds to a coherence length of 83 ± 2 μm .

Then, we fix the position at the point where the visibility is maximized, and proceed with the fine scan using spectral selection with the monochromator (see inset in Figure 4.5). Note that the period of the fringes corresponds to the wavelength of the idler photons, which agrees with the theoretical prediction (see the second Equation in (4.3)). Without a sample, the visibility is $14.0 \pm 0.4\%$. The main factor limiting the visibility of the interference fringes is the imperfection of the optical elements. Independent measurements of DM₂ show a reflection of $\sim 20\%$ for IR light (2500 – 2900 nm) and a transmission of $\sim 70\%$ for visible light (650 – 750 nm). The nonlinear crystal has

Fig. 4.6 **Measured spectrum of the signal SPDC photons.** The central line of the spectrum is at $\lambda_s = 694.4 \pm 0.1$ nm, the FWHM is $\Delta\lambda = 5.8 \pm 0.2$ nm. Therefore, the coherence length of the SPDC photons is $l_{coh} = 83 \pm 2$ μm . The dashed line corresponds to the sinc^2 fitting curve with $R^2 = 0.97$. The bottom x axis corresponds to the detected wavelengths of the signal photons λ_s , the upper x axis show the conjugate wavelength of the idler photons λ_i .

the absorption of about $\sim 20\%$. From Equation (4.8) we estimate that the expected visibility is about 15%, which is in agreement with our experiment. Note that further improvement in the visibility is possible by optimization of coatings and materials of the optical elements.

From the change of the visibility of the inference fringes, we infer the transmission coefficient (see Figure 4.7). In a similar way, we collect the data at different wavelengths. The tunability is achieved by rotating the crystal and selecting the signal photon wavelength by the monochromator. The data is collected at 1 nm steps for the signal photons, which corresponds to a step of 10 nm for the idler photons.

The obtained transmission spectrum of the PDMS sample is shown in Figure 4.7 (a), and is in a good agreement with the measurement of the same sample with a commercial IR spectrophotometer (Shimadzu UV-3600 Plus).

We apply the same measurement procedure for BK7 glass, and AR coated and non-coated silicon samples (with 1 mm thickness each). The measured transmission spectra are shown in Figures 4.7 (b), (c) and (d), respectively. The two sets of data are in good agreement, which proves the validity of our technique.

A relatively thick sample in the idler arm introduces a significant optical delay, which should be compensated for by translating the mirror in the signal/pump arm. Mirror translation may lead to misalignment of the interferometer since the focus of the spherical mirror in the signal/pump beam is displaced from the SPDC crystal. Thus, to recover the interference fringes after the insertion of thick samples, we also introduce a well-characterized glass plate in the signal/pump arm which compensates the optical thickness of the sample.

Our measured refractive indices are in good agreement with the literature (see Table 4.1) [110–112]. The accuracy of the refractive index measurements is $\Delta n_i^m = 0.02$, and accuracy of the transmissivity is $\Delta T_i^m = 0.02$. Reflection and absorption

Fig. 4.7 Transmission spectra of (a) a PDMS sample with 143 μm thickness, (b) BK7 sample with a 1 mm thickness, (c) silicon sample without AR coating, with 1 mm thickness, (d) silicon sample with AR coating, with 1 mm thickness. The blue squares show the experimental results obtained by our method; the red curves show the data obtained using a commercial IR spectrophotometer. The top axis indicates the actual measured wavelength of signal photons and the bottom axis indicates wavelength of idler photons. Error bars in (a)–(d) show the standard deviation.

coefficients are calculated using Equation (4.8) taking into account the obtained refractive index and transmission coefficient data, see Table 4.2. The accuracy of the reflection coefficient measurement is $\Delta R_i^m = 0.003$ and the accuracy of the absorption coefficient measurement is $\Delta \alpha_i^m = 0.06 \text{ cm}^{-1}$.

The accuracy of the measurements of the absorption coefficient is determined by the uncertainty of the measurements of the interference visibility. Due to the high count rate of signal photons (tens of kHz) the relative uncertainty in the measured visibility is small. This enables accurate and precise measurements of the absorption coefficient even at relatively low visibility values.

The spectral range of our technique is defined by the transparency range of the LiNbO₃ crystal ($0.4 - 5 \text{ } \mu\text{m}$). We can further extend the range to mid- and far-IR range by an appropriate choice of nonlinear crystals. For instance, an AgGaS₂ (AGS) crystal would allow extending the transparency range up to $12 \text{ } \mu\text{m}$. Since the SPDC is a routine technique used for generating THz radiation, we expect that our technique is also applicable to this range [113].

4.4 Summary

In this chapter we demonstrated a method for measuring optical constants in the mid-IR range by detecting the interference of visible photons using a nonlinear Michelson interferometer. We obtain the refractive index, transmission, reflection and absorption coefficients of samples in a broad spectral range with good accuracy and spectral resolution.

Our technique is applicable to samples opaque in the visible range, which is highly relevant to characterization of optical materials for applications in IR-photonics, telecom, and material analysis. We also eliminate the requirement of prior information about the sample properties at the visible range, which is a critical limitation of our earlier

work. The obtained results are in good agreement with data taken using conventional IR spectroscopy techniques and literature data [110–112]. The accuracies of our measurements of the refractive index ($\Delta n_i^m = 4 \cdot 10^{-2}$) and transmission coefficient ($\Delta T_i^m = 2 \cdot 10^{-2}$) are on par with state-of-art refractometers and ellipsometers.

Sample thickness, μm	Wavelength, nm	Refractive index, n_i^m		Transmittance, T_i^m	
		Our method	Database	Our method	Database
PDMS (143 $\pm$ 2)	2200	1.4 $\pm$ 0.02	1.41	0.93 $\pm$ 0.02	0.93
BK7 (980 $\pm$ 10)	2600	1.51 $\pm$ 0.03	1.48	0.87 $\pm$ 0.02	0.88
Si w/o AR (1030 $\pm$ 10)	2200	3.50 $\pm$ 0.07	3.45	0.49 $\pm$ 0.04	0.52
Si w AR (1050 $\pm$ 10)	2200	3.50 $\pm$ 0.07	3.45	0.95 $\pm$ 0.02	0.94

Table 4.1 Values of the refractive index and transmittance (mean $\pm$ s.d.) of the samples calculated from the measurements shown in Figures 4.5, 4.7 and using Equations (4.6), (4.7).

Sample	Wavelength, nm	Reflectance, R_i^m		Absorption coefficient, $\alpha_i^m, \text{cm}^{-1}$	
		Our method	Database	Our method	Database
PDMS	2200	0.030 $\pm$ 0.003	0.029	0.67 $\pm$ 0.06	0.69
BK7	2600	0.041 $\pm$ 0.004	0.038	0.57 $\pm$ 0.06	0.51
Si w/o AR	2200	0.31 $\pm$ 0.01	0.30	0.00 $\pm$ 0.05	0.00
Si w AR	2200	0.025 $\pm$ 0.005	0.30	0.00 $\pm$ 0.01	0.00

Table 4.2 Calculated reflectance and absorption coefficients (mean $\pm$ s.d.) of the samples using Equations (4.8).

Chapter 5

Infrared optical coherence tomography and infrared imaging

5.1 Method

In this chapter we further extend the use of the nonlinear interference and realize a method of optical coherence tomography (OCT), where the reflecting layers of the sample can be studied. In this method, the wavelength of the probing photons is tuned to IR range to minimize the scattering, while the wavelength of the detected photons is set in the detection-friendly visible region.

We develop the technique of nonlinear interference to demonstrate imaging through an opaque substrate, measurements of the optical thickness and the sample birefringence in a broad IR range, all in the same measurement configuration. Our approach is based on the nonlinear Michelson interferometer described in Chapter 4, which is versatile and straightforward to implement. So far, none of the previously reported schemes have achieved the same level of functionality in a single configuration.

The scheme of the method is shown in Figure 5.1. Frequency non-degenerate, collinear SPDC field is generated by a pump beam in a nonlinear crystal [26]. The

Fig. 5.1 **Scheme of the nonlinear Michelson interferometer for IR OCT measurements.** Arrows indicate the SPDC k-vectors; dashed lines show their trajectories. The laser generates SPDC field in the nonlinear crystal. Then, SPDC field is split by a dichroic mirror (DM) into visible and IR arms. The pump beam reflected by reference mirror M also generates SPDC field (s_2 , i_2). Interference of SPDC is detected as a function of the displacement of the mirror M. The sample is inserted at the path of the idler photons in the interferometer. Properties of the sample in IR range are inferred from the interference of photons in visible range. Green, orange, and red arrows represent the pump, signal and idler photons, respectively. The beams are shifted for clarity.

phase matching conditions are chosen in such a way, that the wavelengths of the signal (detected) and the idler (probe) photons are in the visible and IR range, respectively. A dichroic mirror DM separates SPDC field. Signal and pump beams are reflected by a reference mirror M and the idler beam is reflected by the surfaces/interfaces of the sample under study. SPDC field is also generated from the second pass of the pump through the crystal.

Let τ_i^m be the amplitude transmission coefficient of the sample, and r_i^m be the amplitude reflection coefficient of the internal interface. Then, similarly to the section 4.1, we can define the following annihilation operator (see Figure 5.2) [24, 25, 59]:

$$a_{i2} = e^{i\phi_i} \left(\tau_i^{m2} r_i^m a_{i1} + \tau_i^m r_i^m \sqrt{1 - \tau_i^{m2}} a_{i0'} + \tau_i^m \sqrt{1 - r_i^{m2}} a_{i0''} + \sqrt{1 - \tau_i^{m2}} a_{i0'''} \right), \quad (5.1)$$

where $a_{i0'}$, $a_{i0''}$, $a_{i0'''}$ describe vacuum fields entering from unused ports of the beam splitter with wavevector k_i . In Equation (5.1) the first term describes the propagation of idler photons through the sample and their reflection by the internal interface. The

sample properties (transmittance, reflectivity and refractive index) at the wavelength of the idler photons can be inferred from measurements of the interference pattern of the signal photons. Direct detection of the idler photons is not required. When the interferometer arms are balanced ($\Delta t = 0$), the visibility of the interference pattern is given by:

$$V = \frac{P_s^{max} - P_s^{min}}{P_s^{max} + P_s^{min}} = |r_i^m| |\tau_i^m|^2. \quad (5.3)$$

For a sample with N reflecting surfaces, the interference signal can be found at several positions of the reference mirror M. The corresponding visibility values are given by:

$$V_n = |r_n^m(z_n)| \prod_{k=0}^{n-1} |\tau_k^m|^2 (1 - |r_k^m|^2), n \in [1, N], \quad (5.4)$$

where r_k^m is the amplitude reflection coefficient from the k -th boundary at the depth z_k , τ_k^m is the amplitude transmission coefficient determined by the absorption and scattering inside the sample, and the last term ($\prod_{k=0}^{n-1} \dots$) corresponds to losses due to reflection from the previous $(n - 1)$ surfaces. Thus, from the measurements of the interference visibility for the signal photons we determine the reflection coefficients of the sample for the idler photons. From the separation between positions of the mirror M, where the interference is observed, we infer the optical thickness of the sample. The axial resolution (optical thickness resolution) of our method is defined by the width of the correlation function $|\mu(\Delta t)|$, which depends on the bandwidth of SPDC photons [39].

5.2 Experimental setup

The experimental setup is shown in Figure 5.3. The pump is a continuous wave (CW) laser (Nd:YAG at 532 nm, 80 mW or Ar+ at 488 nm, 34 mW), which is reflected by the dichroic mirror DM₁. The lens F₁ ($f = 200$ mm) focuses the pump into the

periodically poled Lithium Niobate (PPLN) crystal (length $L = 1$ mm), where SPDC occurs. Lenses F' ($f = 75$ mm) form collimated pump, signal and idler beams after the crystal [37, 38]. Using collimating lenses we no longer need to insert a compensator in the visible arm, to account for the optical delay introduced in the IR arm of the interferometer (like we did in Chapter 4). However, in this case there is the chromatic dispersion of the lenses.

The SPDC field propagates collinearly and is separated into two arms of the interferometer by the dichroic mirror DM_2 . A silver coated mirror M_s , which is mounted on a motorized translation stage, reflects signal SPDC field and the pump beam. A

Fig. 5.3 Experimental setup using a nonlinear Michelson interferometer for IR OCT measurements. A CW laser is used as a pump, and it is output through the dichroic mirror DM_1 . The SPDC field is produced collinearly in the PPLN crystal. SPDC field is separated into two channels by the dichroic mirror DM_2 . Lenses F' collimate the beams. The interference of signal photons is observed as a function of the position of the mirror M_s either by an APD or a CCD camera. Lens F_2 collimates the signal beam. The noise is filtered by the dichroic mirror (DM_1), and by notch (NF) and bandpass (BP) filters. The sample is placed into the IR arm of the interferometer. Mirror M_i is used for calibration of the method. In the imaging configuration, the additional lens F is introduced into the IR arm of the interferometer.

sample under test is inserted in the path of idler photons. In our experiment we use several samples: a Silicon window (Edmund Optics), a compound retardation waveplate (Thorlabs) and a Chromium coated microscope resolution target test (Negative 1951 USAF microscope target on glass substrate, ThorLabs). In the absence of a sample, the gold-coated mirror M_i is used as a reference.

The reflected pump beam generates another SPDC field in the PPLN crystal. We scan the position of the mirror M_s and observe the modulation of the intensity of the single signal photons using a Silicon avalanche photodiode (APD) for data acquisition (Perkin Elmer, AQR-14FC) or a CCD camera (ThorLabs) for setup alignment. Using a PPLN crystal with high SPDC generation efficiency enables us to use an inexpensive CCD camera. This allows one to decrease further the cost of the method. The obtained interference patterns by the CCD camera are shown in Figure 5.4. The beams are overlapped efficiently, if the collimating lenses F' are placed at the focal distance from the crystal (see Figure 5.4 (a)). When one of the lenses F' is misaligned, the interference pattern starts to have several fringes (see Figure 5.4 (b)).

We use a single off-the-shelf PPLN crystal (Covesion) with nine inscribed regions of different poling periods. The wavelength of the SPDC photons is chosen via the

Fig. 5.4 Interference pattern in the spatial domain obtained by the CCD camera when (a) the collimating lenses F' are placed at the focal distance f from the crystal, and (d) one of the lenses F' is misaligned.

selection of the poling period (Λ_{PPLN}) and the crystal temperature (T_{PPLN}). For this demonstration we perform experiments at four different wavelengths of the idler photons (see Table 5.1). The spectra of the detected photons are measured using a home-built grating spectrometer. The spectrum of the SPDC photons defines the coherence length of the signal, which is given by $l_{\text{coh}} = \frac{\lambda^2}{\Delta\lambda}$.

For the raster imaging of the microscope resolution target test, we insert an additional imaging lens F ($f = 50$ mm) in the path of the idler photons. The focal plane of the lens is positioned at the sample surface. For each position of the sample, we measure the visibility of the interference by scanning the mirror M_s . By translating the sample in the transverse plane (x, y) with respect to the incident beam we reconstruct its lateral image.

Probe photon wavelength λ_i (FWHM), nm	Detected photon wavelength λ_s (FWHM), nm	Pump wavelength λ_p , nm	Coherence length l_{coh} , μm	PPLN temperature T_{PPLN} , K	PPLN poling period Λ_{PPLN} , μm
1543 (29)	812.2 (8.2)	532	80 ± 2	399	7.4
2140 (24.5)	707.9 (2.7)	532	185 ± 3	465	8.36
2504 (35)	606.1 (2.0)	488	184 ± 3	399	7.4
3011 (43)	582.4 (1.61)	488	211 ± 3	465	8.03

Table 5.1 Characteristics of the spectra of the SPDC photons in the IR optical coherence tomography experiment.

5.3 Results

5.3.1 Measurements of the positions of reflective layers

We perform optical coherence tomography measurements at four different wavelengths as shown in Figure 5.5. Table 5.1 shows the characteristics of the obtained

spectra. Here, we use $\lambda_p = 488$ nm to obtain longer probe (idler) wavelengths, as it is only possible to generate wavelengths up to $\lambda_i = 2140$ nm with $\lambda_p = 532$ nm.

In Figure 5.5, the width of the spectra decreases with the wavelength due to the phase matching conditions. Narrowing of the bandwidth of the SPDC spectrum leads to an increased coherence length. The spectral shape may not have the form of the sinc^2 function, since they have been modified using band-pass filters in the experimental setup to reduce the noise from the environment at other wavelengths.

First, we perform the experiment with probing photons at $\lambda_i = 1543$ nm (idler) and detected photons at $\lambda_s = 812$ nm (signal). For system calibration, we measure the intensity modulation of the signal photons without the sample. We insert the gold mirror M_i in the IR arm and scan the position of the mirror M_s . Once the

Fig. 5.5 (a)-(d) Spectra of the signal SPDC photons in the optical coherence tomography experiment. Spectra are obtained with band-pass filters in the setup.

optical paths of the signal and the idler photons are equalized within the coherence length of the SPDC, we observe the modulation of the intensity of signal photons (see Figure 5.6 (a)). We chose this point as an origin for the mirror M_s displacement ($z = 0$ mm). The visibility $V_{ref} = 81 \pm 1\%$ of the interference pattern is measured from the fine scan of the mirror M_s (see Figure 5.7 (a)). Note that, in accordance with Equation (5.2), the modulation period of interference fringes for signal photons is given by the wavelength of idler photons.

Then, we insert the sample and repeat the scan (see Figure 5.6 (b)). We use an uncoated pure Silicon window (Edmund Optics) with a thickness of $1080 \pm 4 \mu\text{m}$. The absorption and scattering coefficients for the sample are negligible at the probing wavelengths. Thus, assuming a non-absorptive sample with $N = 2$ surfaces, the visibility ratios are given by (see Equation (5.4)):

$$\begin{aligned} \frac{V_1}{V_{ref}} &= \frac{|r_1^m(z_1)|}{|r_{ref}^m|}, |\tau_0^m| \equiv 1, |r_0^m| = 0, \\ \frac{V_2}{V_{ref}} &= \frac{|r_2^m(z_2)|(1 - |r_1^m(z_1)|^2)}{|r_{ref}^m|}, \end{aligned} \quad (5.5)$$

where $r_1^m(z_1)$, $r_2^m(z_2)$ are the amplitude reflection coefficients by front and back surfaces of the sample respectively, and r_{ref}^m is the amplitude reflection coefficients of the reference mirror M_i .

The detected interference signals at $z_1 = -10.1$ mm and $z_2 = -6.2$ mm correspond to the reflection of idler photons from the front and back surface of the Silicon window, respectively. The interference observed at $z_{12} = -2.3$ mm corresponds to the secondary reflection of idler photons from the two surfaces of the Silicon window. Its expected visibility is $V_{12} = V_2|r_1^m(z_1)||r_2^m(z_2)|$. The visibility for the signal reflected from the back surface and multiple reflections is less than for the signal reflected from the front surface due to the loss of idler photons (see Equation (5.5)).

Fig. 5.6 The intensity of the signal photons detected by the APD versus the position of the mirror M_s . Results in (a) and (b) are obtained for probing photons at $\lambda_i = 1543$ nm (detected photons at $\lambda_s = 812$ nm); in (c) and (d) at $\lambda_i = 2140$ nm ($\lambda_s = 708$ nm); in (e) and (f) at $\lambda_i = 2504$ nm ($\lambda_s = 606$ nm); in (g) and (h) at $\lambda_i = 3011$ nm ($\lambda_s = 582$ nm). (a), (c), (e) and (g) are signals obtained with the reference mirror M_i ; (b), (d), (f) and (h) are signals obtained with the Silicon window sample when IR photons are reflected from different surfaces of the sample.

Figure 5.6 (c), (e) and (g) show measurement results with the reference mirror M_s with probing idler photons at $\lambda_i = 2140$ nm (detected signal at $\lambda_s = 708$ nm), $\lambda_i = 2504$ nm (detected signal at $\lambda_s = 606$ nm), and $\lambda_i = 3011$ nm (detected signal at $\lambda_s = 582$ nm), respectively. The visibility of the interference is measured from the fine scan of the mirror M_s (see Figure 5.7 (b), (c) and (d)). They are equal to $V_{ref} = 33 \pm 1\%$ at 2140 nm, $V_{ref} = 20 \pm 1\%$ at 2504 nm and $V_{ref} = 18 \pm 1\%$ at 3011 nm. These visibilities are somewhat lower, than in Figure 5.7 (a), mainly due to imperfections of optical components in the IR arm. The transmission coefficient of the IR photons through the dichroic mirror is 95% at $1.54 \mu\text{m}$, 70% at $2.14 \mu\text{m}$, 72% at $2.5 \mu\text{m}$, and 66% at $3 \mu\text{m}$ wavelengths. The AR coating of the lens F' is optimized at $1.55 \mu\text{m}$ (reflection is about 0.1%), but for longer wavelengths, the reflection is about 4%. The AR coating on the PPLN crystal provides 5% reflectivity at $1.54 \mu\text{m}$, 8% reflectivity at $2.14 \mu\text{m}$, and 9.5% at $2.5 \mu\text{m}$ (unknown for $3 \mu\text{m}$).

We can compare the visibilities of the interference patterns at $1.54 \mu\text{m}$ and $2.14 \mu\text{m}$ wavelengths, since the same pump laser is used in these measurements. From the above parameters of the optical components, the ratio of interference visibilities at $1.54 \mu\text{m}$ and at $2.14 \mu\text{m}$ is estimated to be 0.60, while the experimental value is 0.41. The difference can be attributed to the reduced spatial overlap of SPDC modes at $2.14 \mu\text{m}$ due to the chromatic dispersion of the lenses [69].

The experimental results for the Silicon sample with probing photons at 2140 nm, 2504 nm and 3011 nm are shown in Figure 5.6 (d), (f) and (h), respectively. Similar to the measurement at 1543 nm, the interferograms show reflections from the front, back and multiple surfaces of the sample. As expected, the visibility of the signal reflected from the back and multiple surfaces is decreased due to the gradual loss of idler photons.

Fig. 5.7 **Fine scans of the interference fringes shown in Figure 5.6 (a), (c), (e) and (g).** The dependence of intensity of the signal photons on the position of the mirror M_s are obtained for probing photons at **(a)** $\lambda_i = 1543$ nm ($\lambda_s = 812$ nm); **(b)** $\lambda_i = 2140$ nm ($\lambda_s = 708$ nm); **(c)** $\lambda_i = 2504$ nm ($\lambda_s = 606$ nm); **(d)** $\lambda_i = 3011$ nm ($\lambda_s = 582$ nm). Points indicate the experimental data; solid curves show the cosine fitting of the data according to Equation (5.2). Note that the modulation period is given by the wavelength of the idler photons λ_i .

From Equation (5.5) we calculate reflection coefficients of the Silicon window at all four probing wavelengths (see Table 5.2). Corresponding refractive indices are calculated using Fresnel equations. Our results are in good agreement with the database values [111]. As expected, the refractive index for Silicon decreases with increasing wavelength of the probing photons. The achieved accuracy of the reflectance measurements is $\Delta R_i^m = 0.003$, and the accuracy of the refractive index is $\Delta n_i^m = 0.014$.

Wavelength λ_i , nm	Measured reflectance, R_i^m	Database reflectance, R_i^m [111]	Measured refractive index, n_i^m	Database refractive index, n_i^m [111]
1543	0.306 ± 0.003	0.306	3.476 ± 0.014	3.478
2140	0.302 ± 0.010	0.303	3.441 ± 0.025	3.448
2504	0.287 ± 0.015	0.302	3.31 ± 0.16	3.440
3011	0.286 ± 0.015	0.301	3.30 ± 0.16	3.433

Table 5.2 Calculated reflectance R_i^m and refractive indices n_i^m of the Silicon window.

5.3.2 Measurement of a birefringence of a sample with hidden features

To test our technique with a more sophisticated sample, we carried out measurements with a compound zero-order retardation waveplate (Thorlabs). By performing measurements we found that the waveplate consists of two quartz plates with orthogonal axes with $934 \pm 9 \mu\text{m}$ and $953 \pm 9 \mu\text{m}$ thickness, respectively. There is an air gap between the plates with $123 \pm 9 \mu\text{m}$ thickness. It is designed to introduce half wavelength retardation at 532 nm. The waveplate is inserted into the idler arm of the interferometer with its fast optical axis parallel to the polarization of idler photons.

The interference pattern without the sample is shown in Figures 5.6 (a) and 5.7 (a), where the probing wavelength is 1543 nm, and the detected wavelength is 812 nm. The interference fringes with the waveplate is shown in Figure 5.8. Signals at $z_1 = -7.16$ mm and $z_4 = -4.11$ mm correspond to reflections from the front and back surfaces of the waveplate, respectively. Two additional peaks at $z_2 = -5.72$ mm and $z_3 = -5.60$ mm correspond to reflections from internal interfaces of the compound waveplate. From the optical delay between reflections #1 and #2 ("front"- "internal 1") we calculate the refractive index along the fast axis (n_o). From the optical delay between reflections #3 and #4 ("internal 2"- "back") we calculate the refractive index along the slow axis (n_e).

The calculated values are shown in Table 5.3 and they are in a good agreement with database values [111].

Fig. 5.8 The intensity of the signal photons detected by the APD versus the position of the mirror M_s with the inserted compound waveplate. The idler (IR) photons are reflected from multiple surfaces of the waveplate. Four interference peaks correspond to reflections from the front (#1), two internal (#2, and #3) and the back interfaces (#4). An optical delay between reflections #1 and #2 gives the fast refractive index (n_o) and the optical delay between reflections #3 and #4 gives the slow refractive index (n_e).

Wavelength λ_i , nm	Measured refractive index, n_o	Database refractive index, n_o [111]	Measured refractive index, n_e	Database refractive index, n_e [111]
1543	1.56 ± 0.04	1.528	1.57 ± 0.04	1.536

Table 5.3 Calculated refractive indices of the compound quartz waveplate at 1543 nm.

5.3.3 Raster imaging of a sample through a Silicon layer

To show the imaging capability of our technique, we insert a lens in the path of the IR photons and carry out raster imaging of the negative microscope test slide preceded by the AR-coated Silicon window (see Figure 5.9). The probing wavelength is $\lambda_i = 1543$ nm, and the detected wavelength is $\lambda_s = 812$ nm. The IR photons are reflected by different regions of the sample, and we measure the interference pattern for visible photons.

From a raster scan of the sample, we reconstruct its reflectance pattern for IR photons (see Figures 5.9 (a) and (b)). As expected, the measured reflectivity of the Chromium film at 1550 nm is about 63% [111]. Once the probe beam hits the region without Chromium coating (number "4" in Figure 5.9 (a) and the vertical stripes in Figure 5.9 (b)), we observe almost zero reflectivity. The width of the stripes of the "4" character in Figure 5.9 (a) is $60\text{ }\mu\text{m}$. The reflectivity dose not drop rapidly, as the probing beam size of $\Delta x = 50\text{ }\mu\text{m}$ is comparable to the feature size. The width of the test stripes in Figure 5.9 (b) is $88\text{ }\mu\text{m}$, and they are well resolved.

Fig. 5.9 **Infrared imaging of a sandwiched sample with a microscope test slide preceded by a Silicon window.** (a),(b) High reflectance regions (green) correspond to a chrome coated pattern; low reflectance regions (blue) correspond to the glass substrate. (c) and (d) show the white light microscope images of the sample without and with the Silicon window, respectively. The dashed rectangles in (c) highlight the measured regions in (a) and (b). The scale bar in (c) and (d) is $200\text{ }\mu\text{m}$.

The image of the sample under the white light microscope is shown in Figure 5.9 (c). The dashed squares show the regions of the resolution test slide measured in our experiment. The sample cannot be seen under visible light, once the Silicon window is inserted in front of it (see Figure 5.9 (d)). The greenish color appears due to the AR coating of the Silicon window.

5.3.4 Wide field infrared imaging

As discussed in section 2.3.1, nonlinear interference of the SPDC photons allows us to perform wide field imaging at the IR range by detecting the visible photons [72]. The main idea of the imaging configuration is to shape the angular spectrum of the SPDC and to put the object in the k -space imaging plane as shown in Figure 2.6. The same principle can be applied to perform IR wide field imaging in the nonlinear Michelson interferometer by introducing the $4f$ -imaging configuration.

Previously, in section 5.3.3, we performed the raster IR imaging. In the experiment, the pump beam was focused inside the nonlinear crystal. Therefore, the divergence of the SPDC photons was determined by the divergence of the pump beam. In that case, two lenses focused the SPDC signal into a point.

Here, in order to perform a wide field imaging, the pump beam should be approximately a plane wave. Thus, with only the natural divergence of the SPDC photons, focusing of the signal by a lens leads to k -space imaging of the SPDC photons in the focal plane.

We use a plane wave pump and built a special lens configuration in the interferometer arms to perform IR wide field microscopy, where the probing wavelength of the idler photons is set at $1.54\text{ }\mu\text{m}$, and the detected wavelength of the signal photons at 812 nm . We use two samples (provided by the "K&S" company) for the wide field imaging experiment: an uncovered silicon substrate with the metal contacts on top

(see Figure 5.10 (a)); two bonded silicon chips with metal contacts inside the sample (see Figure 5.10 (b)). Basically, the first sample is the bottom part of the second one.

Figures 5.10 (c)-(f) show the first test results of the wide field IR imaging using our technique. Figures 5.10 (c) and (e) show the reflection image of the first sample at different magnifications. Figures 5.10 (d) and (f) show the reflection image of the sandwiched sample (with the Si layer on top) for the same magnification as in Figures 5.10 (c) and (e), respectively.

The color scale is related to the reflectivity of the sample. The IR photons are mostly scattered from the metal contacts. Therefore, in the images we recognize the dark stripes as the metal contacts.

Fig. 5.10 **Wide field infrared imaging.** (a) and (b) are the silicon chips with metal contacts used for the wide field IR imaging. (c), (d) and (e), (f) are the resulting IR reflection images of the samples. The dashed ovals indicate approximately the same region of imaging.

For the imaging of the interface between two slabs of the second sample we move the interferometer arm M_s to compensate for the optical delay incurred at the top Silicon layer. As it was mentioned previously, the probing wavelength is $1.54 \mu\text{m}$, which is transmitted through the Silicon layer without absorption. Similarly, the dark lines correspond to the metal contacts. In this images, previously separated metal contacts are connected with each other now (see Figure 5.10 (d) and (f)). The achieved axial resolution of the method is $\Delta x = 35 \mu\text{m}$ in Figures 5.10 (c) and (d) and $\Delta x = 28 \mu\text{m}$ in Figures 5.10 (e) and (f).

5.4 Discussion

The axial resolution of the technique is defined by the first order correlation function of the SPDC given by the Fourier transform of the SPDC spectrum (see Equation (2.26)), which is a sinc²-function in our case [26, 48]. As a result, the interference patterns in Figure 5.6 have triangular shapes. Even though the demonstrated resolution is on the order of tens of microns, it can be reduced by at least one order of magnitude using dedicated methods for the generation of broadband SPDC photons. The methods include, but are not limited, using several SPDC crystals [114] or crystals with chirped poling period [115–117], inhomogeneous heating [118] or applying an electric field [44] to the SPDC crystal. The resolution of the OCT is determined by the width of the first-order correlation function, which is inversely proportional to the width of the spectrum. Therefore, one has to broaden the width of the spectrum of the SPDC to achieve the higher spatial resolution in OCT.

Broadening of the spectrum of the SPDC can be achieved, for example, by using several nonlinear crystals with slightly different orientations of the optical axis (or tilted relative to each other). However, in such a configuration possible gaps between nonlinear crystals may affect the phase relation between generated photons and pump

photons. Therefore, the broadening of the SPDC spectrum may become inhomogeneous, which doesn't result in the narrowing of the correlation function.

Another method of the broadening of the SPDC spectrum is based on application of temperature or electrical field gradient across the nonlinear crystal. In this case, different parts of the crystal have slightly different refractive indices due to the thermo- or electro-optical effects, respectively. The superposition of the SPDC signals generated at the different parts of the crystal forms the broadband spectrum.

The most common method of the broadening of the SPDC spectrum is by using periodically poled nonlinear crystals. The generation of a broadband SPDC spectrum is achieved by using chirped crystals, where the periodicity of the poling increases linearly along the crystal length. The first-order correlation function is determined as the Fourier transform of the spectral intensity. Therefore, the efficient narrowing of the first-order correlation function can be achieved. In the paper of M. B. Nasr et al. [115], the authors generate SPDC with an ultra-broadband spectrum of 300 nm, with the corresponding width of the correlation function of about 2 μm .

Moreover, the smooth spectral shape of the SPDC is beneficial for OCT in comparison with some classical broadband sources (for example Silicon Carbide rod [119]), which is essential for providing high-quality interferograms and better signal-to-noise ratio [120]. The spatial resolution of the imaging in the lateral plane can be further improved by using focusing lenses with short focal distances or microscope objective lenses.

The central wavelength of the probing beam can be set to the required value by an appropriate choice of the nonlinear crystal and the wavelength of the pumping laser. The operation range of wavelengths of the method is limited by the absorption of the nonlinear crystal in the IR range (up to 5 μm for the Lithium Niobate). It can be readily extended into the mid- and far-IR ranges by changing the nonlinear crystal

and the optics. For instance, by using an AGS crystal, it is possible to generate idler photons up to $\lambda_i = 12 \mu\text{m}$.

The sensitivity to the refractive index change is determined by the accuracy of the measurement of the interference visibility. From the results in Table 5.3 we calculate that the technique is capable of resolving the refractive index change at the air/sample interface of $\Delta n_i^m = 0.01$. Higher sensitivity can be archived by careful optimization of the optical losses and the overlap of the beams.

We note that some of the earlier experiments on 2D imaging [72] and optical sectoring [121] used a nonlinear Mach-Zehnder interferometer. However, the scheme based on a Michelson interferometer presented in this section is much more straightforward and versatile. Indeed, it uses only one nonlinear crystal, which is easier to align and to tune the wavelengths. It also requires fewer optical elements such as lenses and mirrors.

5.5 Summary

In this chapter we demonstrated a proof-of-concept of a tunable IR OCT technique, where information about the sample properties at the IR range is inferred from measurements of visible photons. We demonstrate the operation of the technique at four wavelengths: 1543 nm, 2140 nm, 2504 nm, and 3011 nm with the actual detection at 812 nm, 708 nm 606 nm and 582 nm, respectively. The tuning between the wavelengths is rather seamless and requires only the selection of the poling period, the temperature of the crystal and the pump wavelength.

The method shows good precision in determining the position of reflecting layers ($\Delta z = 82 \mu\text{m}$), refractive indices ($\Delta n_i^m = 0.01$) of the sample and its birefringence. In the raster imaging configuration, the technique allows us to perform imaging through a Silicon layer. The resolution ($\Delta x = 50 \mu\text{m}$) and the tuning range can be further improved by a judicious choice of optical elements, such as nonlinear crystals, beam

splitters, lenses, etc. Further study of the impact of scattering can help in extending the operation range of the proposed OCT technique to the mid- and far-IR.

We also demonstrate a proof-of-concept for wide field IR imaging, where the probing wavelength is at $1.54\text{ }\mu\text{m}$ and the detected wavelength is at 812 nm . The achieved axial resolution of the method is $\Delta x = 28\text{ }\mu\text{m}$, which can be further improved by introducing a lens system with higher magnification.

Practical applications of the technique include (but are not limited to) analysis of highly scattering materials in mid-IR for defectoscopy and for the photonics industry [122]. Imaging through Silicon layers and thickness analysis is highly relevant to the microelectronics industry as it enables using accessible photodetectors and light sources for imaging through Silicon wafers.

Chapter 6

Polarization properties of the nonlinear interference and infrared polarimetry

Moving towards the practical implementation of the IR spectroscopy and imaging, one needs to assess the influence of various experimental factors on the performance of the technique. The influence of the SPDC spatial modes overlap [62, 69], the SPDC temporal properties [61], and the losses in the system [59] on the visibility of the interference fringes have been widely studied since the first demonstration of the phenomenon. However, our present understanding of the influence of polarization properties of the down-converted photons on the nonlinear interference is rather limited [123–127].

The paper of T. P. Grayson et al. [124] demonstrates that the interference of signal photons experience a non-local effect of a Pancharatnam phase, which is introduced into the path of idler photons. In their experimental demonstration the idler photons propagate through a set of two quarter wave plates twice, in the forward and backward directions. The first waveplate is oriented at 45° with respect to the initial polarization

of idler photons, and second waveplate is at an angle φ . After the forward pass the initially linear polarization of the idler photons is set at an angle 2φ . Then, at the backward pass the polarization of the idler photons is restored. Despite the fact that the final polarization of the idler photons is the same, the nonlinear interference of signal photons acquires so called the Pancharatnam phase shift due to the rotation of the polarization.

In recent paper of M. Lahiri et al. [126] authors use the nonlinear Mach-Zehnder interferometer for the observation of nonlinear interference, and introduce a polarizer into the path of the signal photons and an attenuator into the path of the idler photons to demonstrate that partial polarization phenomenon may be not always explained using a classical description.

In this work we show that the modification of the polarization of the idler photons changes both the phase and the visibility of the interference pattern of the signal photons. These studies can be applied in IR polarimetry measurements, IR imaging [72, 89], IR OCT (see Chapter 5), to a quantum gate [128] and for quantum entanglement [129, 130].

6.1 Method

The nonlinear interferometer is shown in Figure 6.1 (a). The setup was considered in detail in Chapters 4 and 5 (see Figures 4.1, 4.2, 5.1 and 5.2). According to the beam-splitter model [58], when the idler modes $i1$ and $i2$ are aligned and the sample under study is inserted into the path of the idler photons, the photon annihilation operator for the idler mode $i2$ is given by (see Figure 6.1 (b)):

$$a_{i2,\sigma} = e^{i\phi_i} \left(\tau_i^2 a_{i1,\sigma} + \tau_i \sqrt{1 - \tau_i^2} a_{0',\sigma} + \sqrt{1 - \tau_i^2} a_{0'',\sigma} \right), \quad (6.1)$$

where $a_{0',\sigma}$ and $a_{0'',\sigma}$ stand for vacuum fields entering from the vacuum ports of the beam splitters, σ is the polarization of the photons, τ_i is the complex amplitude transmission coefficient of the beam splitter for the idler photons, and ϕ_i is the phase acquired by the mode $i1$ due to propagation in the interferometer.

Similar to Equation (2.26), the count rate for the signal photons is given by:

$$P_s(\Delta t) \propto 2 \left(1 + |\tau_i|^2 |\mu(\Delta t)| \cos \left(\phi_p - \phi_s - \phi_i + \arg(\tau_i^2) + \arg(\mu(\Delta t)) \right) \right), \quad (6.2)$$

$$\mu(\Delta t) = \int |F(\Omega)|^2 \exp(-i\Omega\Delta t) d\Omega,$$

where $\mu(\Delta t)$ is the normalized correlation function of the SPDC [24], and Δt is the time delay between the propagation times of signal $s1$ and idler $i1$ photons in the interferometer, Ω is the detuning frequency [48] (see Equation (2.14)).

Fig. 6.1 Scheme of the nonlinear Michelson interferometer with a birefringent sample. (a) The pump beam generates SPDC field ($s1$, $i1$) at the first passage through the nonlinear crystal. The SPDC field is split by the dichroic mirror into different arms, and then recombined at the crystal by the mirrors M_s and M_i . Another SPDC field ($s2$, $i2$) is generated by the pump beam at the second pass through the nonlinear crystal. The birefringent sample is inserted into the path of the idler photons. (b) The scheme of the beam splitter model for the birefringent sample in the Michelson interferometer. The idler SPDC photons propagate twice through the sample with the amplitude transmission coefficient τ_i . Mode a_{i1} transforms into mode a_{i2} by introduction of modes $a_{0'}$ and $a_{0''}$ from the vacuum ports of the beam splitter. The crucial point here is that the mirror M_i flips the Cartesian coordinate system (xyz) from right to left.

In Equation (6.2) we consider the overall transmission through the sample τ_i . Let us now determine τ_i in Equation (6.2) taking into account the polarization properties of the sample.

We consider a retardation waveplate as the sample under study. We use the Jones matrix formalism [131, 132] and introduce the corresponding transformation matrix T_{wp} , which is given by:

$$T_{wp} = \tau_i^m \begin{pmatrix} e^{i\delta'/2} & 0 \\ 0 & e^{-i\delta'/2} \end{pmatrix}, \quad (6.3)$$

where $\delta' = \frac{2\pi(n_e - n_o)L^m}{\lambda_i}$ is the relative phase difference between the extraordinary and the ordinary waves acquired by the probe wavelength λ_i in the waveplate of thickness L^m , and τ_i^m is the transmission through medium accounting for reflection, absorption and scattering losses.

Then, the rotation of the Cartesian coordinate system before and after the waveplate is taken into account:

$$J = R(\varphi)T_{wp}R(-\varphi),$$

$$R(\varphi) = \begin{pmatrix} \cos \varphi & -\sin \varphi \\ \sin \varphi & \cos \varphi \end{pmatrix}, \quad (6.4)$$

where $R(\varphi)$ is the rotation matrix, and φ is the angle between the initial coordinate system and the coordinate system along the optical axis of the waveplate (see Figure 6.1 (b)).

Accounting for a double pass of the idler photons through the waveplate and reflection by the mirror M_i , the Jones matrix for this system is given by $J' = R(\varphi)T_{wp}MT_{wp}R(\varphi)$, where M is the reflection matrix of the mirror M_i (as shown in Figure 4.2). Considering

all the transformations we can write the following Jones matrix:

$$J' = \tau_i^{m2} \begin{pmatrix} t & r \\ -r^* & t^* \end{pmatrix}, \quad (6.5)$$

where $|t|^2 + |r|^2 = 1$, t and r are the complex amplitude transmission and reflection functions of the waveplate, respectively:

$$\begin{aligned} t &= \cos \delta' + i \sin \delta' \cos 2\varphi, \\ r &= i \sin \delta' \sin 2\varphi. \end{aligned} \quad (6.6)$$

The horizontally polarized (along the x coordinate) idler mode $i1$ in a Jones vector representation is given by vector $e = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ [131, 132]. Propagating through the waveplate the vector is modified as $J' \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \tau_i^{m2} \begin{pmatrix} t \\ -r^* \end{pmatrix}$. The amplitude transmission function of the idler photons to the same mode and polarization is determined by $\tau_i^2 = \tau_i^{m2} t^*$. Therefore, the absolute value of the transmission coefficient is given by:

$$|\tau_i|^2 = |\tau_i^m|^2 |t| = |\tau_i^m|^2 \sqrt{\cos^2 \delta' + \sin^2 \delta' \cos^2 2\varphi}, \quad (6.7)$$

where φ is the angle between the initial polarization of the idler photons and the optical axis of the waveplate.

Then, the count rate for the signal photons is defined as:

$$P_s(\Delta t) \propto 2 \left(1 + |\tau_i^m|^2 |t| |\mu(\Delta t)| \cos \left(\phi_p - \phi_s - \phi_i + \arg(\tau_i^{m2}) + \arg(t) + \arg(\mu(\Delta t)) \right) \right). \quad (6.8)$$

When the polarization of the idler photons is parallel to the fast ($\varphi = 0^\circ$) or to the slow ($\varphi = 90^\circ$) axis of the waveplate, the propagating wave faces the ordinary (n_o) and extraordinary (n_e) refractive indices, respectively. According to the Equation (6.8),

the relative phase shift of the interference pattern in these cases is determined by δ' :

$$\delta' = \phi_i(\varphi = 90^\circ) - \phi_i(\varphi = 0^\circ) = \frac{2\pi(n_e - n_o)L^m}{\lambda_i}. \quad (6.9)$$

By measuring the relative phase shift of the interference fringes δ' it is possible to characterize the waveplate at a given probing wavelength of the idler photons λ_i .

From Equation (6.8) it follows that besides coherence characteristics $|\mu(\Delta t)|$ and losses $|\tau_i^m|^2$ in the system, the retardation properties of the sample also has to be taken into account for the calculation of the visibility of the interference fringes:

$$V = |\tau_i^m|^2 |\mu(\Delta t)| |t| = |\tau_i^m|^2 |\mu(\Delta t)| \sqrt{\cos^2 \delta' + \sin^2 \delta' \cos^2 2\varphi}. \quad (6.10)$$

Thus, the visibility of the interference pattern is proportional to the modulus of the transmission function of the waveplate $|t|$ as well.

From Equation (6.10) it follows that the maximum and minimum visibilities of the interference fringes are observed at $\varphi = 0^\circ/90^\circ$ and $\varphi = 45^\circ$, respectively. Then, their ratio is given by:

$$\frac{V_{min}}{V_{max}} = \frac{\sqrt{\cos^2 \delta'}}{\sqrt{\cos^2 \delta' + \sin^2 \delta'}} = |\cos \delta'|. \quad (6.11)$$

Using Equations (6.9) and (6.11) it is possible to characterize the type of a waveplate at the given wavelength of idler photons. Since these equations account for the double pass of idler photons through the waveplate, in a single pass the waveplate is characterized by $\delta'/2$.

6.2 Experimental setup

The experimental setup shown in Figure 6.2 is similar to the one we had in Chapter 5. It has only small modifications of optical elements and translation stages. Here, a

continuous wave (CW) laser at 532 nm wavelength (Laser Quantum, Ventus) is used as a pump. The pump beam is focused into a PPLN crystal (Covesion, 1 mm length) by a lens F_1 ($f = 200$ mm), where SPDC field is generated. The phase matching condition of the crystal is adjusted to generate signal photons at $\lambda_s = 809.2$ nm and idler photons at $\lambda_i = 1553$ nm wavelength by heating the crystal to $T_{\text{PPLN}} = 403$ K and choosing the poling periodicity of the crystal to be $\Lambda_{\text{PPLN}} = 7.4$ μm . Then the generated SPDC field is separated by a dichroic mirror DM_2 (Semrock) into visible and IR arms, respectively, and collimated by lenses F' (Edmund Optics with AR coating for a broad visible range; ThorLabs with AR coating for the IR range; both $f = 75$ mm). All three beams are reflected back into the crystal by mirrors M_s (silver coated) and M_i (gold coated) forming a nonlinear Michelson interferometer. The reflected pump beam again generates SPDC field.

The interference of the signal photons is observed once the interferometer arms are balanced within the coherence length l_{coh} of the SPDC photons. Therefore, the mirror M_s is put onto a translation stages (PI, M-505.4DG, precision ~ 1 μm) to equalize the interferometer arms (Δz_s). Then, the pump is filtered away by the dichroic mirror (DM_1 , Semrock), the notch filter (NF, Semrock) and the band pass filter (BP, 809.2 nm ± 0.6 nm). The interference of the signal photons is detected by using a collimating lens F_2 ($f = 200$ mm) and APD (PerkinElmer) or CCD camera (ThorLabs) together with lenses F_3 ($f = 50$ mm) and F_4 ($f = 100$ mm), respectively. Here, the CCD camera is used for the alignment of the overlap of the spatial modes.

Several waveplates are used as test samples. The sample is mounted into a rotation stage (ThorLabs) and inserted into the path of the idler (IR range) photons. Then the fine scanning of the interference of signal photons is performed by translating the mirror M_i (Δz_i), which is mounted onto the piezo stage (PI, P-527.3CD, with

Fig. 6.2 **The experimental setup for the polarimetry measurements.** The CW green laser pumps the PPLN crystal, where SPDC occurs. The phase matching condition is chosen to generate one of the photon at the visible range (signal) and another at the IR range (idler). Then the SPDC field is split by the dichroic mirror DM_2 into different arms (visible and IR) at the Michelson interferometer. Then, all beams are returned back into the crystal by the mirrors M_s and M_i . The filters DM_1 , NF and BP are used to filter out the pump beam from the signal. The interference is detected in two ways: using multimode fiber and the APD detector or by the CCD camera. The mirror M_s is put onto a translation stage for the optical path length adjustment Δz_s , when the sample is inserted into the path of idler photons. The mirror M_i is placed onto a piezo-translator for the measurement of the fine interference fringes.

~ 2 nm precision). The interference fringes are measured for different orientations of the waveplate φ , as shown in Figure 6.2 by an arrow.

6.3 Results and discussion

First, the described experimental setup is tested using waveplates with known properties. We use quarter- (QWP) and half- (HWP) waveplates designed for 1550 nm wavelength. The probing wavelength of the idler photons is chosen to be at $\lambda_i = 1553$ nm,

while the detected photons are at $\lambda_s = 809.2$ nm. At the beginning the interference fringes are measured without the waveplate in the IR arm as a reference (see Figure 6.3). Then, the waveplate is inserted into the IR arm with its optical axis parallel to the polarization of the idler photons ($\varphi = 0^\circ$). The change of the optical path length is compensated for by the translation of the mirror M_s . At the optimized position the measurements are performed by translating the mirror M_i using the piezo-stage to get the fine interference fringes with nanometer precision. The same measurements are done for different orientations of the waveplate φ .

For the HWP@1550nm, QWP@1550nm, HWP@532nm (Thorlabs) the phase shifts are approximately the same, as they have close optical thicknesses. For the QWP@532 nm (DayOptics) the phase shift is less.

Figures 6.4 (a) and (b) show the interference fringes of the signal photons measured for the orientation of the waveplate at $\varphi = 0^\circ, 45^\circ$ and 90° , where points correspond to the experimental data and the solid curves correspond to fitting by cosine function

Fig. 6.3 Shift of the interference pattern due to additional phase acquired in the waveplate inserted to the path of the idler photons. HWP@1550nm, QWP@1550nm, HWP@532nm and QWP@532nm waveplates with the orientation of the optical axis at $\varphi = 0^\circ$ are used in the experiment. The orientation of the waveplates at $\varphi = 0^\circ$ insures that there is no impact to the polarization of the initial state.

using Equation (6.8). As expected from Equation (6.9), the phase shift δ' of the interference pattern between $\varphi = 0^\circ$ and $\varphi = 90^\circ$ are π and 2π for QWP@1550 nm and HWP@1550 nm, respectively, because of the double pass of the probing idler photons through the waveplate (see Table 6.1).

Figure 6.4 (c) shows the visibility of the interference pattern versus the orientation of the waveplate φ . In accordance with Equation (6.10) the interference fringes have maximum visibility at the orientation of waveplates at $\varphi = 0^\circ$ and 90° , while at $\varphi = 45^\circ$ the visibility reaches its minimum. For the QWP@1550 nm at $\varphi = 45^\circ$ the visibility is nearly zero ($V = 0.005 \pm 0.005$). Solid lines in Figure 6.4 (c) correspond

Fig. 6.4 Count rate of the signal photons at $\lambda_s = 809.2$ nm in the nonlinear Michelson interferometer versus the translation of the reference mirror and the visibility of the interference fringes for different orientations of the waveplates φ . The experiment is performed for (a) QWP@1550 nm and (b) HWP@1550 nm waveplates for their orientation of the optical axis at $\varphi = 0^\circ$, 45° and 90° . Points correspond to the experimental data and the solid lines correspond to the cosine fitting function according to the Equation (6.8) with $R^2 > 0.99$. The relative phase shift between the interference patterns at $\varphi = 0^\circ$ and $\varphi = 90^\circ$ is shown as δ' . (c) The visibility of the interference fringes versus the orientation φ of the waveplate designed for 1550 nm wavelength. Points correspond to the experimental results: the circular points correspond to HWP@1550 nm, the triangular points correspond to QWP@1550 nm; the solid curves show the fitting using Equation (6.10) and accounting for the losses in the interferometer (with the $R^2 > 0.99$). The inset shows the zoomed results for QWP@1550 nm near zero visibility.

Fig. 6.5 **Count rate of the signal photons at $\lambda_s = 809.2$ nm versus the translation of the mirror M_i .** The experiment is performed for (a) QWP@532 nm and (b) HWP@532 nm waveplates with their orientation of the optical axis at $\varphi = 0^\circ$, 45° and 90° . Points correspond to the experimental data and the solid lines correspond to the cosine fitting function according to the Equation (6.8) with $R^2 > 0.99$. The relative phase shift between interference patterns at $\varphi = 0^\circ$ and $\varphi = 90^\circ$ is shown as δ' . (c) The visibility of the interference fringes depending on the orientation φ of the waveplate designed for 532 nm wavelength. Points correspond to the experimental results: the circular points correspond to HWP@532 nm, the points with triangle shape correspond to QWP@532 nm; the solid curves show the fitting using Equation (6.10) with $R^2 = 0.98$.

to the theoretical curves for the waveplates designed for 1550 nm, where losses in the interferometer are also taken into account. The inset shows the zoomed results of visibility values near $\varphi = 45^\circ$. Using this data and Equation (6.11) the parameter $\delta'/2$ of the waveplate is calculated (see Table 6.1).

The same measurements are done for the half- and quarter- waveplates designed for 532 nm wavelength. As shown in Figures 6.5 (a) and (b), HWP@532 nm and QWP@532 nm operate as a waveplates with arbitrary phase shifts for the probing beam at 1553 nm. Based on Equation (6.9), the calculated birefringence values are shown in Table 6.1. Similarly, Figure 6.5 (c) shows the dependence of the visibility versus the orientation of the waveplate for the HWP@532 nm and the QWP@532 nm. The ratio

between the maximum and minimum values of the visibility of the interference pattern determines the waveplate birefringence according to Equation (6.11) (see Table 6.1).

The accuracy of the birefringence measurement in the proposed IR polarimetry method depends on the configuration of the system. For the phase-shift measurements the accuracy is high and constant $\Delta\delta' = 0.006\pi$. It mainly depends on the instrumental error of the piezo stage. In this type of measurements the interferometer should be well stabilized.

The visibility measurements do not require the phase stabilization. Here, the accuracy of the method is determined by the contrast between the minimum V_{min} and the maximum V_{max} visibilities achieved in the experiment. Better contrast leads to higher accuracy. In this type of measurements we observed $\Delta\delta' = 0.03\pi - 0.002\pi$ accuracy.

These measurements can contribute to the IR polarimetry and IR OCT of samples with an optical activity. The polarization-sensitive optical coherence tomography (PS-OCT) [133, 134] and polarization-sensitive quantum optical coherence tomography (PS-QOCT) [135] have already been set forth as methods for the birefringence measurement of a layered sample, providing an enhanced contrast of the method. Similarly, our technique can be used for the PS-OCT development, extending the conventional methods to the mid-IR range.

The obtained results can also find application in a wide-field imaging [136]. Firstly, the demonstrated technique allows to control the phase of the interference pattern, enabling the introduction of the phase shift π by using a QWP waveplate instead of moving the position of the piezo stage. This increases the stability of the method. Secondly, it allows one to enhance the signal-to-noise ratio of the method by compensating for the change of polarization of the probing light due to an optical activity of the sample.

Waveplate type	Phase shift at 1553 nm $\delta'/2$ (single path), π	
	Visibility method	Phase shift method
HWP@1550 nm	0.980 ± 0.030	1.004 ± 0.006
QWP@1550 nm	0.498 ± 0.002	0.495 ± 0.006
HWP@532 nm	0.321 ± 0.006	0.322 ± 0.006
QWP@532 nm	0.173 ± 0.010	0.172 ± 0.006

Table 6.1 Results for the phase shift measurements for the waveplates probed at $\lambda_i = 1553$ nm wavelength.

6.4 Summary

In this chapter we studied the polarization properties of the nonlinear interferometry technique, where the change of the polarization of the idler photons affects both the visibility and the phase of the interference fringes of the signal photons. The method is demonstrated for several waveplates, which were probed with the idler photons at 1553 nm. Using the visibility and the phase shift measurements we achieve the high accuracy of the method, enabling us to characterize the birefringence of waveplates with uncertainties as low as $\Delta\delta' = 0.002\pi$. The obtained results form the basis for further incorporation of the method into polarimetry, imaging and PS-OCT techniques working in a broad IR range.

Chapter 7

Conclusion and future work

7.1 Conclusion

In this work we have demonstrated the technique of infrared metrology using visible light based on the nonlinear interference of the SPDC photons. In this technique SPDC photons are generated in nonlinear crystals in a strongly frequency non-degenerate regime, where one photon is generated at the visible range (signal) and another in IR range (idler). We use different nonlinear interferometers, where the interference of the signal photons also depends on amplitudes and phases of the idler photons, which are used to probe properties of the medium under study. Therefore, detecting the interference pattern of visible photons allows us to infer the properties of the medium in IR range.

In Chapter 3 we have demonstrated IR spectroscopy measurements of CO₂ gas. We have used the degenerate Mach-Zehnder interferometer to study the refractive index and absorption coefficient of the CO₂ near the strong absorption line at 4.27 μm . Our technique showed an accuracy of the refractive index measurement as $\Delta n_i^m = 5 \cdot 10^{-6}$, and the absorption coefficient measurement as $\Delta \alpha_i^m = 0.02 - 0.2 \text{ cm}^{-1}$ with a spectral resolution of 2.2 cm^{-1} at 2330 cm^{-1} (4.3 μm).

Then, in the same Chapter, we proposed a method to increase the signal-to-noise ratio of the technique by removing the spectrograph and the slit from the detection system. A dedicated image processing algorithm allowed us to increase the signal-to-noise ratio up to eight times due to the detection of the whole interference pattern of the SPDC photons in the transverse plane. In this configuration the method showed an accuracy of $\Delta n_i^m = 5 \cdot 10^{-6}$ in a measurement of the refractive index, and $\Delta \alpha_i^m = 0.05 \text{ cm}^{-1}$ in a measurement of the absorption coefficient. The spectral resolution of the method in this case is about 80 cm^{-1} .

The main advantage of the degenerate configuration of the interferometer is that mechanical perturbations do not affect to measurement results, since all the photons travel along the same path in the interferometer. However, the interference pattern is affected by all the three photons (pump, signal and idler), which interact with the sample. Therefore, a priori knowledge of properties of the sample in the visible range is needed.

In Chapter 4 we have theoretically and experimentally studied the nonlinear Michelson interferometer. This configuration allowed us to split signal (visible) and idler (IR) photons into different paths of the interferometer. Thus, it was possible to put the sample under study into the path of idler photons only. Therefore, the a priori knowledge of sample's properties at the visible range was not needed. We have demonstrated the IR spectroscopy measurements. The method showed good accuracy in measurement of the refractive index ($\Delta n_i^m = 0.02$) and absorption coefficient ($\Delta \alpha_i^m = 0.06 \text{ cm}^{-1}$). The spectral resolution of the method was 5 cm^{-1} at 4550 cm^{-1} ($2.2 \text{ }\mu\text{m}$).

In Chapter 5 we used the nonlinear Michelson interferometer for the IR optical coherence tomography and IR imaging measurements. We used several samples to show a proof-of-concept measurements of the technique, including a Silicon window,

a compound retardation waveplate, a resolution test target and a Silicon chip. The method showed a fair axial ($\Delta z = 82 \mu\text{m}$) and lateral resolution ($\Delta x = 50 \mu\text{m}$) for the raster IR imaging and lateral resolution ($\Delta x = 28 \mu\text{m}$) for the wide field IR imaging. These characteristics can be further improved by an optimized choice of the optical components.

In Chapter 6 we have demonstrated the application of the concept to the IR polarimetry. Using the nonlinear Michelson interferometer we were able to characterize the birefringence of the samples with an accuracy up to $\Delta\delta' = 0.002 \pi$.

Table 7.1 shows an overview of the demonstrated methods, illustrating how the optimal interferometer configuration depends on the task. For the refractive index and absorption coefficient measurements the Mach-Zehnder interferometer shows the best results because of the intrinsic stability of the configuration. However, this configuration needs a priori knowledge about the sample in the visible range.

The method with a nonlinear Michelson interferometer does not need any a priori knowledge about the sample and shows multi-functionality of the measurements in a single configuration. Using this interferometer it is possible to probe the reflectivity, the transmission and the polarization properties of the sample.

In summary, we demonstrated that our technique has distinctive advantages over the conventional IR measurement methods:

1. The wavelength of the probing photon (idler) is in the IR range, while the wavelength of the detected photon is in the visible range. Thereby, we eliminate necessity of IR sources and detectors, reducing the overall cost of the technique;
2. Wide tunability of the SPDC wavelengths in the near- and mid-IR range allows us to investigate sample properties in a broad spectral range;
3. High precision measurements of both the refractive index and absorption coefficient can be made simultaneously and independently;

Nonlinear interferometer type	Degenerate Mach-Zehnder	Degenerate Mach-Zehnder, without spectral selection	Michelson
Refractive index, Δn_i^m	$5 \cdot 10^{-6}$	$5 \cdot 10^{-6}$	0.02
Absorption coefficient, $\Delta \alpha_i^m \text{ cm}^{-1}$	0.02 – 0.2	0.05	0.06
Reflectivity, ΔR_i^m	$\times$	$\times$	$3 \cdot 10^{-3}$
Transmission, ΔT_i^m	$\checkmark$	$\checkmark$	0.005 – 0.02
Polarimetry, $\Delta \delta'$	$\times$	$\times$	0.002π
Raster imaging resolution	$\times$	$\times$	50 μm - lateral 82 μm - axial
Wide field imaging resolution	$\times$	$\times$	28 μm - lateral
Spectral resolution (at λ_i), cm^{-1}	2.2 (2330)	80 (2330)	5 (4550)
Wavelength tunability	Yes	Yes	Yes
Intrinsic stability	Yes	Yes	No
A priori knowledge about visible range	Needed	Needed	Not needed

Table 7.1 Overview of the performance of the demonstrated IR metrology schemes.

4. Our method can be applied to the characterization of various types of samples: gases, liquids, solids, nanoparticles, etc.

We believe that our method can complement, or possibly substitute, conventional IR measurement techniques.

7.2 Future work

Our technique is based on the nonlinear interference of the SPDC photons, which was first proposed several decades ago. Despite the fact that quite a lot of work has already been done on this topic, there are many interesting applications that are yet to be explored.

Range of wavelengths

As was mentioned previously, the demonstrated method is limited by the transparency range of the nonlinear crystal. The transparency range of the Lithium Niobate (LiNbO_3) crystal used in our experiments is from 500 nm up to 5 μm . However, there are many other nonlinear crystals, which can be used for SPDC generation in mid-, far-IR and THz ranges. For example, AgGaS_2 and HgGa_2S_4 crystals have transparency range from 0.5 to 13 μm ; GaSe - from 0.65 to 18 μm , Te - from 3.8 to 32 μm [42, 137]. Moreover, Lithium Niobate crystal can be also used to generate THz range photons [138, 139]. Therefore, it would be interesting to try to widen the range of probe wavelengths to access a broader variety of spectroscopic features.

Stimulated coherence

In our work we studied spontaneously generated photon pairs from parametric down conversion. However, it is possible to use an additional laser at the wavelength of

the idler photons to enter into the stimulated regime of the parametric down conversion (PDC), which increases the generation rate of the photon pairs. Thus, the measurement time of the method can be significantly reduced.

Moreover, in this regime the visibility of the interference pattern is given by $V = \frac{2|\tau_i|}{1+|\tau_i|^2}$, instead of $V = |\tau_i|$ in spontaneous regime [89]. Therefore, the method is less sensitive to losses acquired in the sample (see Figure 7.1). This can enhance the signal-to-noise ratio of the method and can be used to probe highly scattering or absorbing samples.

High gain regime of the PDC generation

In our theoretical framework we considered only the low gain regime of the SPDC generation. However, there are works, where the nonlinear interference in a high gain regime was considered [140–143]. The technique was realized using nonlinear Mach-Zehnder interferometer. Using the nonlinear Michelson interferometer can further increase the functionality of the method. Moreover, realization of the technique in the high-gain regime would lead to a higher signal-to-noise ratio of the method too.

Fig. 7.1 Visibility of the nonlinear interference versus the amplitude transmission $|\tau_i|$ of a sample for the stimulated (pink) and spontaneous (blue) parametric down conversion.

Other types of nonlinear interferometers

In our work we considered two types of the interferometers: Mach-Zehnder and Michelson. However, there are many other types of interferometers, which possibly can be applied in our technique. First of all, it is interesting to observe interference in a cascade system of nonlinear crystals [144]. Potentially, using many crystals can enhance the contrast of the interference pattern. Moreover, it can improve the resolution of the spectroscopy measurements, since the interference fringes become narrower, as was proposed by D. N. Klyshko [74]. Secondly, using a Sagnac interferometer might be interesting in terms of the stability of the method. Similar to the degenerate Mach-Zehnder interferometer, the Sagnac interferometer also has high intrinsic stability due to the symmetry of the system.

List of Publications, Conferences & Awards

Journal Papers

1. **Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Tunable optical coherence tomography in the infrared range using visible photons, *Quantum Science and Technology* **3**(2), 025008 (2018)
2. **Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Measurement of infrared optical constants with visible photons, *New Journal of Physics* **20**, 043015 (2018)
3. **Anna V. Paterova**, SHAUN LUNG, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Nonlinear infrared spectroscopy free from spectral selection, *Scientific reports* **7**, 42608 (2017)
4. **Anna V. Paterova**, SHAUN LUNG, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Infrared spectroscopy assisted by entangled photons, *Proceedings of SPIE* **10030**, 100300X-1 (2016)

5. DMITRY A. KALASHNIKOV, **Anna V. Paterova**, SERGEI P. KULIK, LEONID A. KRIVITSKY, Infrared spectroscopy with visible light, *Nature Photonics* **10**, 98–101 (2016)

One manuscript is under preparation:

- **Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Polarization effects in nonlinear interference of down-converted photons, (2018)

Conference proceedings

1. **Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Infrared metrology using visible photons, *CLEO*, ATh3O.3 (2018)

Conferences

1. **Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Infrared metrology using visible light, *1st Advances in Quantum Engineering International Meeting*, Singapore (2018)
2. **Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Infrared metrology using visible photons, *CLEO Laser Science to Photonic Applications*, San Jose, USA (2018)
3. *Invited talk* **Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Infrared metrology using visible photons, *Institute of Physics Singapore (IPS) Meeting 2018*, Singapore (2018)

4. **Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Infrared metrology using visible photons, *CQT10*, Singapore (2017)
5. **Anna V. Paterova**, SHAUN LUNG, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Nonlinear infrared spectrometer without spectral selection, *CLEO-Pacific Rim 2017*, Singapore (2017)
6. *Invited talk* **Anna V. Paterova**, SHAUN LUNG, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Spectral selection free infrared spectroscopy, *Institute of Physics Singapore (IPS) Meeting 2017*, Singapore (2017)
7. **Anna V. Paterova**, SHAUN LUNG, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Spectral selection free infrared spectroscopy, *Conference on 90 Years of Quantum Mechanics*, Singapore (2017)
8. **Anna V. Paterova**, SHAUN LUNG, DMITRY A. KALASHNIKOV, SERGEI P. KULIK AND LEONID A. KRIVITSKY, Quantum infrared spectrometer, *25th Annual International Laser Physics Workshop*, Yerevan, Armenia (2016)
9. **Anna V. Paterova**, SHAUN LUNG, DMITRY A. KALASHNIKOV, SERGEI P. KULIK AND LEONID A. KRIVITSKY, Infrared spectroscopy with visible light, *International Conference on Quantum Communication Measurement and Computing (QCMC)*, Singapore (2016)
10. **Anna V. Paterova**, DMITRY A. KALASHNIKOV, SERGEI P. KULIK AND LEONID A. KRIVITSKY, Infrared nonlinear spectroscopy without spectral selection, *Institute of Physics Singapore (IPS) Meeting 2016*, Singapore (2016)

11. **Anna V. Paterova**, DMITRY A. KALASHNIKOV, SERGEI P. KULIK AND LEONID A. KRIVITSKY, Quantum spectroscopy, *Single Photon Workshop 2015*, Geneva, Switzerland (2015)

Awards

- **Best Poster Presentation - Anna V. Paterova**, HONGZHI YANG, CHENGWU AN, DMITRY A. KALASHNIKOV AND LEONID A. KRIVITSKY, Infrared metrology using visible photons, *1st Advances in Quantum Engineering International Meeting* (2018)
- **Singapore International Graduate Award (SINGA)** SCHOLARSHIP FOR INTERNATIONAL GRADUATE STUDENTS FOR PHD RESEARCH, SINGAPORE (AUGUST 2014 - AUGUST 2018)
- **Singapore International Pre-Graduate Award (SIPGA)** SCHOLARSHIP FOR INTERNATIONAL UNDERGRADUATE STUDENTS AS PART OF A DIPLOMA WORK, SINGAPORE (OCTOBER - DECEMBER 2013)

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Appendix A

Calibration of the rotation stage

In Chapter 2 we made the treatment for the phase matching condition of the SPDC (see Equation (2.13)). It was mentioned that the phase matching condition in a bulk crystal depends on the refractive index of the crystal at the pump wavelength $n_p(\theta_p)$, where θ_p is the phase matching angle of the crystal. In the "IR spectroscopy without spectral selection" experiment in Chapter 3 (see section 3.2), the phase matching condition was controlled by tilting the crystal, which was placed onto the rotation stage. In the experiment, the calibration of the phase matching angle θ_p versus the tilting angle of the rotation stage γ was done to automate the scanning procedure.

When the pump beam is perpendicular to the crystal, there is no refraction of the incident beam (see Figure A.1 (a)). In this case the phase matching condition is defined taking into account the cut angle of the crystal θ_c and the drift angle inside the crystal $\rho(\theta_p)$ (see Figure A.1 (b)), which is given by [42]:

$$\rho(\theta_p) = \text{Arctan} \left(\left(\frac{n_o}{n_e} \right)^2 \tan \theta_p \right) - \theta_p, \quad (\text{A.1})$$

where n_o and n_e are ordinary and extraordinary refractive indices of the nonlinear crystal.

Fig. A.1 **Propagation of the pump beam inside the nonlinear crystal.** (a) The pump beam is perpendicular to the crystal, (b) The drift angle of the extraordinary wave inside the nonlinear crystal, (c) The pump beam propagates at an angle γ with respect to the crystal.

When the crystal is tilted to an angle γ , we need to take into account Snell's law of refraction (see Figure A.1 (c)):

$$\sin \gamma = n_p(\theta_p) \sin \beta, \quad (\text{A.2})$$

where β is the angle between the propagation of the pump inside the crystal and the z axis. In this case, the phase matching angle will be $\theta_p = \theta_c + \beta$.

Then, we can write Snell's law in terms of phase matching angle θ_p :

$$\sin \gamma = n_p(\theta_p) \sin(\theta_p - \theta_c). \quad (\text{A.3})$$

Taking into account the drift angle $\rho(\theta_p)$ in Equation (A.1), Snell's law is given by:

$$\sin \gamma = n_p(\theta_p + \rho(\theta_p)) \sin(\theta_p - \theta_c - \rho(\theta_p)). \quad (\text{A.4})$$

During the calibration procedure we measured the central wavelengths of the SPDC signal photons λ_s depending on the rotation of the stage γ , see Figure A.2 (a). The minus sign of the stage corresponds to the anticlockwise rotation direction. Based on results in Figure A.2 (a) we calculated corresponding phase matching angles, see

Figure A.2 (b). Figure A.2 (b) shows the experimental points for the phase matching angle θ_p versus the rotation of the stage γ and the theoretical curve given by Equation (A.4) for the cut angle $\theta_c = 50.5^\circ$.

Finally, we fit the experimental data point with a linear function (see Figure A.3). We used this calibration function at the automated procedure of measurements.

Fig. A.2 Central wavelength (a) and phase matching angle (b) of the SPDC signal photons λ_s depending on the rotation of the stage α . The points correspond to the experimental data, and the dashed line shows the curve given by Equation (A.4).

Fig. A.3 **Fitting curve of the phase matching angle versus the tilting angle of the stage γ .** The points show the experimental data, and the solid line correspond to the linear fit.

Appendix B

Measurement of reflectivity and transmissivity of the dichroic mirrors

Figure B.1 shows the reflection and transmission measurements of the dichroic beam splitters used in the experiments. Figure B.1 (a) shows the reflection of the dichroic mirror used in the experiment described in Chapter 4. The dichroic mirror was made of the thin gold layer, and idler photons were reflected from the surface of the mirror. The visibility in this case was mainly determined by the reflection of the idler photons from the dichroic mirror. As we can see from Figure B.1 (b) the reflectivity of the beam splitter was $\sim 20\%$ at $\lambda = 2500$ nm. Therefore, the visibility of the interference pattern was significantly reduced.

Figure B.1 (b) represents transmission coefficient measurement for the dielectric dichroic mirror used in Chapters 5 and 6. In this case the visibility of the interference pattern was determined by the transmissivity of idler photons through the dichroic mirror, which was $\sim 95\%$ at $\lambda = 1550$ nm. Therefore, in the experiment we reached a high visibility of the interference fringes.

Fig. B.1 **Measurement of the dichroic mirrors.** (a) The reflectance of the dichroic mirror used in the experiment described in Chapter 4. (b) The transmittance of the dichroic mirror used in the experiment described in Chapters 5 and 6.

Appendix C

Program codes

In this work we used various software to analyze the obtained data. Thus, for the filtering of the two-dimensional interference pattern in Figures 3.11 (a) and (b) we used the following code written in *Python*:

```
import os

import numpy as np

import numpy.fft as fft

import matplotlib.pyplot as plt
from matplotlib.cm import Greys_r

def load_data(filen):
    data = np.loadtxt(filen)
    return np.delete(data, 0, 1)

def remove_noise(data):
    data = fft.fft2(data)
```

```
# Remove cross shaped rectangular segments
data[:, 106:407] = 0
data[106:407, :] = 0

# Remove noise prone areas
data[0:10, :10] = data[0:10, -10:] = data[-10:, :10] = data[-10:, -10:] = 0
data[:, 100, 0] = data[0, :, 100] = 0

# Inverse transform
data = fft.ifft2(data).real
data -= np.min(data)

return data

if __name__ == "__main__":
    f_list = [
        n for n in os.listdir("C:\\Users\\...\\Anaconda")
        if n.endswith(".asc")
        and "noise" not in n]

    for f_name in f_list:
        data = load_data(f_name) - load_data("noise.asc")
        data = remove_noise(data)

    new_f_name = "filtered_" + f_name[:-3] + ".csv"
```

```
np.savetxt(new_f_name, data, delimiter = ',')

I = load_data("-5_5.asc") - load_data("noise.asc")

plt.imshow(
#e.data,
I,
#data,
cmap = Greys_r,
#origin = 'bottom',
#extent = extent,
#aspect = 'auto'
)
plt.colorbar()

plt.tight_layout()
plt.show()
```

We obtained the one dimensional data in Figures 3.11 (c) and (d) using the following code:

```
import os
import csv
import numpy as np
import matplotlib as mpl
import matplotlib.pyplot as plt
import matplotlib.font_manager as font_manager
```

```
from matplotlib.cm import Greys_r

X0 = 282
Y0 = 297

"""Working with experimental data"""
#signal = np.loadtxt("filtered_-5_5.csv", delimiter=',')

signal = np.loadtxt("650 nm.asc") - np.loadtxt("635 nm noise.asc")
signal_array = np.array(signal)
#new_array = np.delete(signal_array, 0, 1)
np.savetxt("signal", signal_array)
new_array = np.delete(signal_array, 0, 1)

"""Plotting 2D experimental data"""
#plt.imshow(new_array, cmap = Greys_r)
#plt.show()

def load_data(filen):
    data = np.loadtxt(filen, delimiter=',')
    return data

def angular_expression(array, centre):
    """Adds up the points in an array in an angular sweep about a central
    point"""
    xd, yd = array.shape
```

```
x = np.linspace(0, xd - 1, xd)
y = np.linspace(0, yd - 1, yd)
x, y = np.meshgrid(x, y)
x0, y0 = centre
xs, ys = array.shape
#r = np.sqrt((x-x0)**2 + (y-y0)**2)
r = np.arctan(np.sqrt((x-x0)**2 + (y-y0)**2)*0.016/500)*180/np.pi
return r
```

```
def polar(array, center):
    r = angular_expression(array, center)
    flattened = np.vstack((array.flatten(), r.flatten()))
    unordered = np.transpose(flattened)[np.lexsort(flattened)]
    unordered = np.split(unordered, 512)
    #ordered = np.transpose(np.mean(unordered, 1))
    ordered = np.transpose(np.median(unordered, 1))
    return ordered
```

```
def error_bar(array, center):
    r = angular_expression(array, center)
    flattened = np.vstack((array.flatten(), r.flatten()))
    unordered = np.transpose(flattened)[np.lexsort(flattened)]
    unordered = np.split(unordered, 512)
    #ordered = np.transpose(np.nanstd(unordered, 1))
    ordered = np.transpose(np.std(unordered, 1))
    return ordered
```

```

#print error_bar(new_array, (Y0, X0))
y, x = polar(new_array, (Y0, X0))
yerr, xl = error_bar(new_array, (Y0, X0))

if __name__ == "__main__":
    f_list = [
    n for n in os.listdir("C:\\Users\\...\\Anaconda")
    if n.endswith(".csv")
    and "noise" not in n]
    # print (y)

    np.savetxt("signal_", y, delimiter = ',')
    np.savetxt("signal_X_", x, delimiter = ',')
    np.savetxt("error_", error_bar(new_array, (Y0, X0)), delimiter = ',')

    """Plotting one dimensional graph"""
    title_font = {"fontname":"Times New Roman", "size":"20", "color":"blue", "s
    axis_font = {"fontname":"Calibri", "size":"20"}

    ax = plt.subplot()
    #ax.errorbar(x, y, yerr = yerr, fmt = "o")
    for label in (ax.get_xticklabels() + ax.get_yticklabels()):
        label.set_fontname("Arial")
        label.set_fontsize(20)

```

```

plt.plot(x, y)
plt.scatter(x, y, s=5, c=(1,0,0))

plt.plot(x, y)
plt.scatter(x, y, s=5, c=(1,0,0))

plt.xlabel(r"$\alpha$, degree", **axis_font)
plt.ylabel("counts", **axis_font)
plt.ylim([0,360])
plt.xlim(0, 0.25)
plt.title("Projection graph", **title_font)
plt.show()

```

Then, we perform fitting of the one dimensional interference pattern with Equation (3.11). The program finds the best fitting of the experimental data using the least mean square algorithm.

```

# import matplotlib.pyplot as plt
import multiprocessing as mp
from fitter2 import *
import os

def run_optimisation(fname, show_plot=False):
# data
data = load_data(fname) - load_data("-10 noise.asc")
c_angle = float(fname[:-4].replace("_", ".").replace("+", ""))

e = Experiment(257, 194, 532, 630, 700, 0.015, .001, 1, -5.75, data)

```

```
mask = e.circular_mask(1/12, 11/12)

# GIB PRESSURE
n = e.NSCO_HELPER(300/760)

def phase_matching(a):
# 55 deg crystal
return a*0.4392 + 57.768

def I_helper(parameters, ca, Lm):
# a, b, c = parameters
tau, a, b, c, d, oa = parameters

print("{: <10.8f} {: <10.8f} {: <10.8f} {: <10.5f} {: <10.8f} {: <10.5f} {"
# {: >15.10f}

# theta grid
e = Experiment(260, 200, 532, 630, 700, Lm, .001, oa, ca, data)

tcxy = e.tgrid_from_data()

# Modulation function
# tau = 2.71828*(-a*tcxy - b) + c
# tau = 1
```

```

# 7.69358, 2689.89316, 38.31194/2, 7.90015, 2772.03445, 54.32713/2
n_res = e.resonant_NSCO_HELPER(a, b, 38.31194/2, c, d, 54.32713/2, n=e.NS

# Cubic matrix
depth = 32
tcxy, l = e.tcxy_vs_l(tcxy, e.l_lower, e.l_upper, depth)

# Apply a gaussian visibility function
tau *= 1/((2*3.1415)**.5 * .0022/.3) * 2.71828**(-tcxy[0]**2 /2/ (.0022/.3)

# The l**-2 factor arises due to integrating over lambda as opposed to w
I = e.I_lt(l, tcxy, n, tau, Lm, n_res)*l**-2
I = sp.integrate.simps(I, dx=(e.l_upper-e.l_lower)/depth, axis=0)
I = e.scale(I)

return np.sum((e.data[mask] - I[mask])**2)

pm_angle = phase_matching(c_angle)

init_params = (.05, 7.69358, 2689.89316, 7.90015, 2772.03445, pm_angle)
bounds = (
(0, 1),
(7.69358 - .2, 7.69358 + .2),
(2689.89316 - 200, 2689.89316 + 200),

```

```
# (38.31194/2 - 5, 38.31194/2 +5),
# (0.5, 1.5)
(7.90015 - .2, 7.90015 + .2),
(2772.03445 - 200, 2772.03445 + 200),
(pm_angle - .2, pm_angle + .2)
)

res = opt.minimize(
I_helper,
init_params,
# method='SLSQP',
bounds=bounds,
args=(c_angle, 0.030048770074777836)
# options={
#     'ftol':10**-10
# }
)
# TNC, SLSQP

print("Completed for {}".format(c_angle))

print(res)

return c_angle, res["x"]
```

```
if __name__ == "__main__":
    # out = []

    # pool = mp.Pool(processes=4)

    # f_list = [
    #     n for n in os.listdir()
    #     if n.endswith(".asc")
    #     and "noise" not in n
    # ]
    # out = pool.map(run_optimisation, f_list)

    # pool.close()
    # pool.join()

    out = run_optimisation("-2_75.asc"),
    print(out)

    try:
        print("tau, a, b, c, d, c_angle, pm_angle")
        for c, (t, a, b, c, d, p) in out:
            print("({}, {}, {}, {}, {}), # {}, {}".format(t, a, b, c, d, c, p))
    except Exception:
```

```
for row in out:
    print(row)

# SLSQP with l**-2, corners constrained, old refractive index
[
    (-4.5, 0.019999237304218695, 50.111720008302107, 51.11172),
    (-4.75, 0.019707127772470145, 50.075190614421324, 51.01036),
    (-5.0, 0.018436304838579498, 50.0607609715086, 50.909),
    (-5.25, 0.012700839704067275, 50.05835466795142, 50.80764),
    (-5.5, 0.019739031780495776, 50.058559742537604, 50.70628),
    (-5.75, 0.01590681683794605, 50.073217998667964, 50.60492),
    (-6.0, 0.010275654142765269, 50.051029990388884, 50.50356),
    (-6.25, 0.014498138120365779, 50.061899109932902, 50.4022),
    (-6.5, 0.019820543551402475, 50.041913884867895, 50.30084),
    (-6.75, 0.02, 50.028580347041647, 50.19948),
    (-7.0, 0.015020179577118887, 50.066144914704957, 50.09812),
    (-7.25, 0.015170147257385507, 50.079147541901904, 49.99676),
    (-7.5, 0.010888345500809129, 50.062839025285662, 49.8954)
]
```