Video-Rate Imaging with Undetected Photons

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Imaging and microscopy are some of the most important tools in modern life science for getting new insights into metabolisms or unravelling bio-chemical processes. However, in particular low-light observations outside the visible spectrum are still challenging and a limiting factor. A rugged, label-free quantum imaging system is presented capable of recording at video rate in the visible regime, while illuminating the sample with undetected light of different wavelength. The results pave the way for a field deployable quantum imaging device allowing live-cell imaging in extreme spectral ranges with a minimal photo dose.

1. Introduction

Within the framework of the vast development of quantum technology, quantum-enhanced imaging offers real-world applications beyond classical limitations.\[1\] This reaches from noise-reduced imaging\[2,3\] over entanglement enhanced microscopy\[4–8\] to imaging in extreme spectral ranges.\[9–12\] Especially the latter one offers great potential for applications in life science. There, both ends of the spectrum are highly interesting for imaging purposes. For instance chemical selective imaging needs to be carried out in the mid-infrared, which is the spectral fingerprint region for many substances like lipids, proteins, and DNA.\[13\] However, direct imaging in the mid-infrared is difficult due to the large intrinsic noise of mid-infrared cameras,\[14–16\] which fundamentally limits the achievable resolution for the low excitation powers needed to detect the signal.

Not disturb the analyzed samples. This becomes even more critical considering label-free ultraviolet imaging\[17–19\] where the photon dose the samples are exposed to must be as low as possible. Naturally, the minimum dose is limited by the detection efficiency, which is lower for ultraviolet detectors compared to detectors for the visible spectral range. Hence, finding an imaging technique allowing to have different wavelengths for illumination and detection would be highly beneficial. To this end, several approaches are being pursued, such as frequency up-conversion imaging\[11,12,20\] or ghost imaging.\[9,21–23\] While the former one needs large illumination intensities, limiting its applicability for photo-sensitive biological samples, the latter one still rests upon detecting both wavelengths. Although the detection of the extreme wavelength for sample interaction can be carried out without any spatial resolution, it intrinsically limits the overall efficiency. Contrary to ghost imaging, imaging (and spectroscopy) with undetected photons does not require the detection of both wavelengths in order to obtain the image of the object. In fact, the light interacting with the object is not detected at all. Hence, it opens the door for new applications in life science imaging and provides higher flexibility when designing an imaging system.

In order to make this approach available for real-world applications within the field of biomedical imaging, besides the extreme spectral range, one needs to address the following points: portability, stability, imaging performance, and video-rate.

Here, we present a revised, more evolved version of quantum imaging with undetected photons with respect to applicability for life science. We demonstrate a stable, very compact and robust setup able to capture quantum images with undetected photons at video rate, which brings this quantum-enhanced imaging technique within reach for actual field deployment.

2. Theoretical Background

The imaging technique itself is based on Mandel’s effect of induced coherence.\[30,31\] We pump a nonlinear crystal with a laser beam in order to generate correlated signal and idler light via non-degenerate spontaneous parametric down conversion in the low-gain regime. The crystal is placed in a Michelson-type interferometer as shown in Figure 1.\[32–35\]

In our setup, signal and idler beams have different wavelengths. While the idler beam is used to illuminate the object the signal beam is detected with an EMCCD or with a CMOS camera. Pump light can generate signal and idler photons in the
nonlinear crystal either in forward direction (beam paths b and c), or in backward direction (beam paths e and f) after the pump beam is reflected via path d. The system is arranged in a way that the two possibilities for down conversion result in indistinguishable alternatives for signal and idler in beam paths e and f. The indistinguishability leads to interference that can be observed in the intensity of either beams e (signal) or f (idler). The presence of an object in path c (idler) affects the observable interference in the detected signal beam, although no signal photon passes through the object. In addition, due to the momentum correlation between signal and idler beams, the interference pattern observed with a camera for the signal light in beam e allows to reconstruct spatial features of the object although the signal light never interacted with it.

Assuming identical down-conversion rates in both directions, even phases of the individual interferometer arms and neglecting transmission losses an object with transmission and phase of \(t_{oc}\) and phases of the object in the idler arm c will lead to signal detection probability on the camera in path e of

\[ P_s \sim 1 + t_{oc} \cos(\phi_{oc}) \]  

One can clearly see that interference can be observed in the signal beam outside the interferometer due to an object placed in the idler beam path inside the interferometer. Since the actual idler photons are not detected at all, this technique is referred to as "quantum imaging with undetected photons". A full derivation including all transmission functions and phases in the interferometer arms of pump, signal, and idler as well as uneven down-conversion rates is given in the Supporting Information. The interference visibility is given by

\[ V_s = t_{oc} \]  

and is thus directly proportional to the transmission of the object.

The result of interference in the signal beam although an object is placed in the idler beam path is similar to that obtained in refs. [10, 33]. However, no beam splitter is used in our setup.

It shows that both transmission and phase shift of an object placed in idler beam path c can be recovered by detecting only photons in signal beam path e that never interacted with the object and that can have a different wavelength than the idler light.

In the experiment, this phenomenon is exploited in a spatial multimode scenario. The nonlinear crystal is illuminated with a collimated pump beam. Under the assumption of perfect momentum correlation, one can treat all pairs of correlated spatial modes according to above description. As a result, the interference fringe observed at a selected point on the camera contains the information about the transmission and phase shift of a corresponding point on the object. In other words, an image is obtained.

Note that both transmission and phase-shift of the object at the illumination wavelength can be detected by this method. The spatially dependent transmission of the sample can be directly inferred by measuring the visibility of the interference pattern on the camera, whereas phase shifts can be quantified by determining the relative phase of the interference fringe at different points on the camera [10,30]

3. Experimental Setup

The particular setup used in our experiment exploits a 1 mm × 2 mm × 2 mm periodically poled potassium titanyl phosphate (ppKTP) nonlinear crystal, which is pumped from opposite directions by a continuous-wave laser (90 mW; 405 nm). This causes collinear non-degenerate photon pair generation by type-0 spontaneous parametric down-conversion in either of the two directions (beams e and f or beams b and c). The pump power
is low enough to render the possibility of two or more photon pairs simultaneously being present in the setup negligible. The down-converted photon pairs exhibit a wavelength of \( \approx 910 \text{ nm} \) for the signal and \( \approx 730 \text{ nm} \) for the idler. The specific values can be temperature tuned (see Figure S3, Supporting Information) and selected by a narrow band-pass interference filter (1.5 nm) in front of the sCMOS/EMCCD camera, which detects the signal beam e. Contrary to the general convention, we here call signal the lower frequency photons. This way we are consistent with former works denoting signal the photons detected at the camera.

Compared to systems for quantum imaging with undetected photons exploiting Mach–Zehnder interferometric configurations with two nonlinear crystals, a Michelson interferometric setting with only one crystal has a smaller footprint and needs less optical components. This decreases the complexity of the whole system. Hereby, we achieve the goal of realizing a portable fully enclosed device with dimensions 80 cm \( \times 60 \text{ cm} \times 30 \text{ cm} \), although system integration is not performed yet. In addition, the use of one nonlinear crystal allows avoiding any distinguishability due to not perfectly identical crystals.

4. Results and Discussion

We experimentally demonstrate the capabilities of our setup by placing an amplitude mask in beam path c and recording the constructive and destructive interference image from beam e (see Figure 2). Under the condition of maximum constructive, respectively, destructive interference of the images, one obtains a contrast-enhanced version by simple subtraction. The images were taken at 100 ms acquisition time using an EMCCD camera with a gain factor of 20. Due to subtraction, also the background is drastically reduced improving the signal-to-noise ratio (SNR) from 7.6 dB to 10.7 dB. Imaging further test samples (see Figure 3), reveals that sample features of 90 \( \mu \text{m} \) are clearly resolvable.

Replacing the EMCCD by a sCMOS camera allows to capture images without any gain and better signal-to-noise ratio since the photon flux in the system is high enough. This way, we have been able to record with 20 ms acquisition time, which is already video rate.\[36\] Figures 4 and 5 show a video sequence of the same object moving through the field of view taken with the sCMOS camera at 100 ms and 20 ms acquisition time, respectively, without applying any gain factor. As it is shown in these figures, the smallest feature size of the object used for taking the videos is 86 \( \mu \text{m} \). These features are well resolved, which agrees with the expected resolution limit of 60 \( \mu \text{m} \) considering the optical system, the momentum correlations and the undetected beam wavelength.\[37\] The standard deviation of the dark counts in the camera was around 3.5 and 3.6 gray values both exposure times, which we consider as the dark noise and the minimal difference in the signal modulation needed to distinguish between two levels of transmission. Assuming this, it is possible to differentiate a total transmission changes of 0.8 and 0.2, respectively, at 20 ms and 100 ms exposure time, using our signal modulation obtained from Figures 4 and 5 as reference for a transmission of 1.

We would like to put our results with respect to acquisition time and resolution in comparison to related publications. For the acquisition time we compare to the seminal work of ref. [10]

![Figure 2](image_url)
and two very recent publications, refs. [28] and [38], which exhibit 500 ms, 1000 ms, and 500 ms acquisition time, respectively. Our systems allows to enhance the imaging speed by a factor of five, respectively, ten. With respect to the resolution capabilities, one needs to consider the ratio $R$ of the field of view and the resolution. The higher $R$ the better the resolution capabilities. Considering again the two very recent publications, refs. [28] and [38], field of view to resolution ratios of $R \approx 28$ and $R \approx 65$ can be extracted. Considering that the smallest structures could be easily resolved our setup features a ratio of $R > 58$. Assuming the theoretical limit of 60 $\mu$m resolution results in $R \approx 83$.

The maximum visibility measured with the system is 77% (see Figure 8). For calculating the theoretical maximum visibility, we follow Equation (2), taking into account transmission losses from optical components such as dichroic mirrors (see Equation (8), Supporting Information). The transmission losses ($T_{\text{theo}}$) are calculated based on data sheet values and experimentally verified ($T_{\text{exp}}$) to be ($T = |t|^2$) for the pump $T_{\text{p}}^\text{theo} = 80.7\%$ and $T_{\text{p}}^\text{exp} = (75 \pm 8)\%$, for the signal $T_{\text{s}}^\text{theo} = 95.3\%$ and $T_{\text{s}}^\text{exp} = (92 \pm 4)\%$, and for the idler $T_{\text{i}}^\text{theo} = 86.8\%$ and $T_{\text{i}}^\text{exp} = (85 \pm 5)\%$. With these values into Equation (2), the maximum visibility achievable is 92.8% from the theoretical values, and (92 ± 3)% from the experimentally measured transmission values. In practice, one encounters additional effects that lower the visibility. These include imperfections in the optical components such as chromatic aberrations and the limited precision of the optical system alignment. The decrease in visibility mainly comes from two effects. The first one occurs when the photons of the forward and backward direction become distinguishable. This happens when the imaging system is not perfect. One possible impact is that idler photons of the forward direction pass through the crystal with a spatial mode that is different from the backward one. They are then at least partially distinguishable, which decreases the interference and thus the visibility. The second effect occurs due to spontaneously down-converted photons not being perfectly momentum correlated. For a fixed signal photon momentum, the associated idler photon momenta follow a certain probability distribution. If within the range of this distribution the pass through beam path $c$ produces different phase shifts (or transmission values), the visibility can also be reduced. For a more detailed explanation of this effect see refs. [39, 40]. To estimate the impact of a misaligned optical system, we considered the effects of displacing the lenses and tilting the mirrors in beam paths $b$ and $c$. We start with the case for which all lenses are perfectly placed at their nominal focal length distance from the crystal, that is 150 mm for the lenses used (L1, L2, and L3 in Figure 1). We further assume all path lengths to be adjusted in order to show interference. In this case, we can achieve the maximum visibility of 92.8% limited only by transmission losses. For obtaining the numerical results in Figure 6, we consider misaligned configurations, which are realistic for the adjustment accuracy of the real optical system.

The setup is as follows: the lenses in the signal and idler paths $b$ and $c$ are shifted by 1 mm along the optical axis in the direction of the crystal. While this longitudinal displacement alone does not reduce the visibility much, an additional tilt of the mirrors at the end of the beam paths does. This shows that as long as the mirrors are well aligned the alignment tolerance of the lenses is not critical. The mirror tilt is compensated by a transverse shift of...
Figure 6. Theoretical visibility values related to the tilt of the mirrors in the signal and idler path. The black line marks the configurations where the experimentally observed visibility of 77% is obtained. The maximum visibility of 92.8% due to transmission losses is achieved at zero tilt in both mirrors.

Figure 7. Phase stability of the imaging system. Comparison of the stability achieved without active phase stabilization (grey line) and with active phase stabilization (black line). The two images with the interference fringes show the phase drift during 30 min, which is not perceptible by eye. A longer test, over 24 h, was performed with the active stabilized system, demonstrating an excellent long-term phase stability.

Figure 8. Visibility stability of the imaging system over a 24 h test. Visibility remains constant at 75 ± 2%.

5. Conclusion

In summary, we have developed and implemented a portable quantum imaging setup with undetected photons, able to capture images in video rate. Several aspects of the imaging capabilities have been investigated. The smallest sample feature sizes of 86 µm could be resolved easily. Here, an improved design can enhance the resolution and thus, the applicability for cell imaging in the future. The theoretically analyzed interference visibility matches the experimental measurements. The maximum visibility of 77% can be explained by minor misalignment that lead to an increased distinguishability of photons being generated in forward or backward direction. Further improvement of the visibility is expected by exploiting low loss optical components and superachromatic lenses. We demonstrated that with an active stabilization both the visibility and the phase remain highly stable over long-term measurements. Our results pave the way for in vivo biochemical imaging with undetected photons.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.
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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

imaging with undetected photons, nonlinear interferometers, quantum imaging

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