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# Supplementary Materials for

# Microscopy with undetected photons in the mid-infrared

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## Working Principle

Our imaging setup is based on the conceptual arrangement of Michelson-type nonlinear interferometer (*25*). A pump beam illuminates a nonlinear crystal twice in sequence, in a folded geometry. Photon pairs are formed in the first and second pass through the crystal, the signal and idler are denoted $\left|c\right\rangle_s \left|d\right\rangle_i$ and $\left|e\right\rangle_s \left|f\right\rangle_i$ respectively. After the first path, the idler and signal are split using a dichroic mirror (DM). The idler is sent into a sample with transmittance T and phase shift $\gamma$: $\left|c\right\rangle_s \left|d\right\rangle_i \rightarrow Te^{i\gamma} \left|c\right\rangle_s \left|d\right\rangle_i + \sqrt{1-T^2} \left|c\right\rangle_s \left|l\right\rangle_i$, where the state $\left|l\right\rangle_i$ encompasses all the loss in the idler arm. Both idler and signal are then back reflected and aligned to allow minimal distinguishability between the bi-photon amplitudes ($\left|c\right\rangle \rightarrow \left|e\right\rangle, \left|d\right\rangle \rightarrow \left|f\right\rangle$) such that after the second pass of the crystal, the obtained state is:

$$\left|\phi\right\rangle = \frac{1}{2}[(1+Te^{i\gamma}) \left|e\right\rangle_s \left|f\right\rangle_i + \sqrt{1-T^2} \left|e\right\rangle_s \left|l\right\rangle_i] \tag{1}$$

Subsequently, the idler photon is discarded using a DM and the detection probability obtained is:

$$P = \frac{1}{2}(1 + T\cos\gamma) \tag{2}$$

Accordingly, an interferometric image with visibility T can be detected. The idler is not detected and the information about the object in the idler path is transferred to the signal through the high spatial correlations shared between the signal and idler modes. This feature when combined with non-degenerate downconversion allows sensing and detection at different wavelength ranges.

## Theoretical Model

To ascertain the theoretical imaging capacity of our implementation we developed a simple theoretical framework to calculate both the anticipated field of view (FoV) and resolution of our optical system (specified in full width half maximum (FWHM)). We also make a comparison to the theory of the SPDC wavefunction.

The FoV in our imaging system is impacted by the emission angle of the down-converted idler light and any subsequent magnification that defines the size of the illuminating spot

$$\mathrm{FoV} = \frac{2f\tan(\theta_i)}{M} \approx \frac{2f\theta_i}{M}, \tag{3}$$

where $f$ denotes the focal length of the collimating optical element adjacent to the crystal, $\theta_i$ is the divergence angle (corresponding to the half width half maximum (HWHM)), and $M$

represents the magnification of the optical system after the collimation. The opening angle, $\theta_i$, results from the phase-matching conditions

$$\theta_i = \lambda_i \sqrt{\frac{2.78}{L\pi} \frac{n_i n_s}{\lambda_i n_s + \lambda_s n_i}} \tag{4}$$

where $L$ denotes the crystal length, $\lambda_i$ and $\lambda_s$ are the wavelengths of the idler and signal respectively and $n_i$ and $n_s$ are the reflective indices of KTP at the designated wavelengths.

The resolution is limited by two constraints. The first limitation arises from the standard diffraction limit, applicable to most conventional imaging and microscopy techniques,

$$\delta x_{NA} = \frac{\lambda}{2NA}, \tag{5}$$

where $\lambda$ and $NA$ are the wavelength and numerical aperture of the optical elements in our system.

The second condition upon the resolution - and here the limiting condition - is an expression of the strength of momentum (or position) entanglement inherent to the bi-photon state (*32, 33*) and is given by

$$\delta x_{corr} = \frac{\sqrt{2 \ln 2} f \lambda_i}{\pi w_p M}, \tag{6}$$

where $w_p$ denotes the pump waist. The number of spatial modes is therefore

$$N_{modes} = \left( \frac{\text{FoV}}{\delta x_{corr}} \right)^2 = \frac{5.56\pi}{\ln 2} \frac{{w_p}^2}{L} \frac{n_i n_s}{\lambda_i n_s + \lambda_s n_i} \tag{7}$$

Unsurprisingly, the number of spatial modes does not depend on the magnification ($M$) - provided one has a sufficiently large NA. By substituting the experimental values into Eq.7 we obtain $911 \pm 13$ spatial modes.

## Derivation of the emission angle

The width of the idler in the far field corresponds to an emission angle, $\mathrm{sinc}^2(\frac{\Delta k L}{2}) = \frac{1}{2}$, corresponding to a phase mismatch $\Delta k L = 2.78$. Expressing the phase mismatch in terms of the transverse emission angle we obtain

$$(\frac{\pi n_i}{\lambda_i} {\tilde{\theta}_i}^2 + \frac{\pi n_s}{\lambda_s} {\tilde{\theta}_s}^2) \frac{L}{2} = 1.39. \tag{8}$$

Assuming transverse momentum conservation, the idler emission angle within the crystal is given by

$$\tilde{\theta}_i = \sqrt{\frac{2.78}{\pi L} \frac{n_s {\lambda_i}^2}{n_s n_i \lambda_i + {n_i}^2 \lambda_s}}, \tag{9}$$

corresponding to angle in free space of

$$\theta_i = n_i \cdot \tilde{\theta}_i = \lambda_i \sqrt{\frac{2.78}{\pi L} \frac{n_s n_i}{n_s \lambda_i + n_i \lambda_s}}. \tag{10}$$

## Experimental values

The signal wavelength was measured using a spectrometer to be $\lambda_s = 801 \pm 1nm$. The pump wavelength was measured using a wave meter and the value obtained is: $\lambda_p = 659.75 \pm 0.01nm$. The idler wavelength was calculated according to energy conservation ($\lambda_i = \frac{1}{\frac{1}{\lambda_s} + \frac{1}{\lambda_p}}$), the obtained value is: $\lambda_i = 3.74 \pm 0.02\mu$m. The refractive indexes were calculated using the appropriate Sellmeier equations (*34, 35*) for KTP: $n_s = 1.845, n_i = 1.752$. The length of crystal is: $L = 2$mm and the pump waist at the crystal is $w_p = 431 \pm 6\mu$m.

The SNR was calculated for the difference image, by dividing the number of mean counts by the standard deviation within a 7x7 pixel region. The size of the region was chosen to be comparatively small in order to exclude the effect of the varying illumination profile.

The power on the sample was measured by detecting the signal light on the camera after the first path of the pump through the crystal. The number of counts on the camera was converted to the the number of electrons according to a constant conversion factor (provided by PCO) and then to the number of photons using the quantum efficiency of the CMOS detector at 800 nm ($N_{photons} = \frac{0.46 N_{counts}}{0.42}$). The accordingly obtained number of $1.5 \cdot 10^8$ photons at a wavelength of 3.5 $\mu m$ corresponds to sample illumination power of 17 pW. Given the bandwidth of the photons of around $1.5 \cdot 10^13$ Hz, the brightness could be increased by more than 4 orders of magnitude without leaving the low-gain regime of SPDC. In the high-gain regime on one hand spectral and spatial mode numbers would start to decrease, while on the other hand quantum-enhanced phase-sensitivity effects would become relevant.

## Future setup optimization

A future increase in the crystal aperture from 1 to 4 cm will allow the increase of total spatial modes by $4^2 = 16$, according to Eq. 7. To attain a SNR of 10, while maintaining the same pump power, we plan to optimize the number of pixels per resolvable elements and reduce it by 2 per direction (from 5 down to 2.5), corresponding to a total factor of 4. Also, by improving the visibility for the magnified setup and reaching the unmagnified visibility corresponding to a SNR of 25, we will be able to reduce the current exposure time from 1 to 0.65 seconds ($1sec \cdot \frac{16}{(25/10)^2 \cdot 4}$). For a spectral resolution of 2.5 $cm^{-1}$ and a spectral range of 625 $cm^{-1}$, a hyperspectral image with 250 spectral modes can be recorded in just 2.7 minutes.

## Theoretical formulation from the bi-photon state

In the low gain limit, the bi-photon state produced by the SPDC can be written in the angular spectra representation

$$|\psi_1\rangle = \int C_1(\mathbf{q}_i, \mathbf{q}_s) |\mathbf{q}_s, \mathbf{q}_i\rangle, \tag{11}$$

where $\mathbf{q}_s$ and $\mathbf{q}_i$ represent the transverse components of the SPDC emission. Analogous to the spectral properties of SPDC emission, the spatial properties are governed by the intersection of energy and momentum conservation,

$$C_1(\mathbf{q}_i, \mathbf{q}_s) = \alpha(\mathbf{q}_s, \mathbf{q}_i)\phi(\mathbf{q}_s, \mathbf{q}_i), \tag{12}$$

specified by the $\alpha(\mathbf{q}_s, \mathbf{q}_i)$, the pump function and $\phi(\mathbf{q}_s, \mathbf{q}_{\mathbf{i}})$ is the phase-matching function, respectively. We recast longitudinal phase mismatch $\Delta k_{z_0}$ in terms of the transverse momenta

$$\Delta k_z = \Delta k_{z_0} - \tfrac{1}{2}\left(\tfrac{|\mathbf{q_s}+\mathbf{q_i}|^2}{k_p} + \tfrac{|\mathbf{q_s}|^2}{k_s} + \tfrac{|\mathbf{q_i}|^2}{k_i}\right) \tag{13}$$

where $\Delta k_{z_0}$ is the residual longitudinal collinear phase-mismatch at the given emission wavelength. The phase function is given by

$$\phi(\mathbf{q}_s, \mathbf{q}_i) = \mathrm{Sinc}\left(\frac{L\Delta k_z}{2}\right), \tag{14}$$

and the pump function is

$$\alpha(\mathbf{q}_s, \mathbf{q}_i) = \exp\left(-|\mathbf{q}_s + \mathbf{q}_i|^2\omega_p^2/4\right), \tag{15}$$

where $\omega_p$ is the pump waist. Consider two identical SPDC processes aligned in series with all modes matched

$$|\Psi\rangle = \tfrac{1}{\sqrt{2}}(|\psi_1\rangle + \psi_2\rangle) \tag{16}$$

More explicitly,

$$|\Psi\rangle = \tfrac{1}{\sqrt{2}}\int d\mathbf{q}_s d\mathbf{q}_i C_1(\mathbf{q}_i, \mathbf{q}_s)|\mathbf{q}_i, \mathbf{q}_s\rangle + \tfrac{1}{\sqrt{2}}\int d\mathbf{q}_s d\mathbf{q}_i \exp i\phi\, C_1(\mathbf{q}_i, \mathbf{q}_s)|\mathbf{q}_i, \mathbf{q}_s\rangle \tag{17}$$

If we want to consider an overall transmission, $\eta$, of the idler between the first and second crystal, we considering an additional loss mode $\mathbf{q}_i^{\star}$. Considering an intensity measurement in the Fourier plane, one obtains

$$\langle\Psi|\mathbf{q}_s^{\dagger}\mathbf{q}_s|\Psi\rangle = \frac{1}{2}(\eta + 2\sqrt{\eta}\cos\phi + 1)\int d\mathbf{q}_i d\mathbf{q}_s |C_1(\mathbf{q}_i, \mathbf{q}_s)|^2 \tag{18}$$

If we assume no longitudinal phase mismatch at all wavelengths, then the number of modes decreases linearly with increasing wavelength (where one has chosen to fix the pump wavelength). This is simply a consequence of increasing non-degeneracy reducing the spatial entanglement of the system. For the unmagnified configuration and our earlier specified system values, the conditional probability density gives a resolution of $326\mu$m (FWHM). The theoretical FoV is 9.0 mm (FWHM). The Schmidt decomposition of the joint probability density gives an effective mode number of 500. The discrepancy between the Schmidt number and the mode number obtained earlier, reflects that the latter considers the ratio of a Sinc width to a Gaussian width.

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