

Supplementary Information

High-speed mid-IR imaging: chemically selective 2D and 3D imaging with InGaAs camera

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Conduction electrons generation through NTA process

The detected signals in the NTA experiments are directly proportional to the number of photo-excited electrons in the conduction band. The change in the number of conduction electrons N_c due to the NTA process depends on the loss of the number of $\hbar\omega_1$ and $\hbar\omega_2$ photons, indicated by N_1 and N_2 . For each photo-excited electron, one $\hbar\omega_1$ photon and one $\hbar\omega_2$ photon are simultaneously absorbed. The change in N_c with respect to the propagation depth z in the semiconductor material can thus be written as:

$$\frac{dN_c}{dz} = -\frac{1}{2} \left(\frac{dN_1}{dz} + \frac{dN_2}{dz} \right) \quad (S1)$$

Nonlinear absorption is typically modelled through the coupled equations of the irradiances of the ω_1 and ω_2 beams, which are affected by losses due to degenerate and non-degenerate two-photon absorption processes:

$$\begin{cases} \frac{dI_1}{dz} = -\alpha_{DTA}(x_1)I_1^2 - 2\alpha_{NTA}(x_1, x_2)I_1I_2 \\ \frac{dI_2}{dz} = -\alpha_{DTA}(x_2)I_2^2 - 2\alpha_{NTA}(x_2, x_1)I_1I_2 \end{cases}$$

$$x_1 = \frac{\hbar\omega_1}{E_g}, \quad x_2 = \frac{\hbar\omega_2}{E_g} \quad (S2)$$

Since field irradiance can be related to the photon number as $I = \frac{\hbar\omega}{\pi\tau\omega_0^2}N$, equation S2 can be re-written in terms of change of the number of photons:

$$\begin{cases} \frac{dN_1}{dz} = -2\frac{\hbar\omega_2}{\tau\pi\omega_0^2}\alpha_{NTA}(x_1, x_2)N_1N_2 \\ \frac{dN_2}{dz} = -2\frac{\hbar\omega_1}{\tau\pi\omega_0^2}\alpha_{NTA}(x_2, x_1)N_1N_2 \end{cases} \quad (S3)$$

where τ is the interaction time and $\pi\omega_0^2$ is the illumination area on the detector. Equation (S3) expresses the change in the number of photons due to the non-degenerate absorption processes. Combining equations S1 and S3, the photogenerated careers can be written as:

$$\begin{aligned}
\frac{dN_c}{dz} &= \frac{N_1 N_2}{\tau \omega_0^2} (\hbar \omega_2 \alpha_{NTA}(x_1, x_2) + \hbar \omega_1 \alpha_{NTA}(x_2, x_1)) \\
&= \frac{N_1 N_2 E_g}{\tau \pi \omega_0^2} (x_2 \alpha_{NTA}(x_1, x_2) + x_1 \alpha_{NTA}(x_2, x_1))
\end{aligned}
\tag{S4}$$

Since

$$\begin{aligned}
\alpha_{NTA}(x_1, x_2) &= K \sqrt{E_p} \frac{F_{NTA}(x_1, x_2)}{n_1 n_2 E_g^3} \\
F_{NTA}(x_1, x_2) &= \frac{(x_1 + x_2 - 1)^{3/2}}{2^7 x_1^3 x_2^4} (x_1 + x_2)^2,
\end{aligned}
\tag{S5}$$

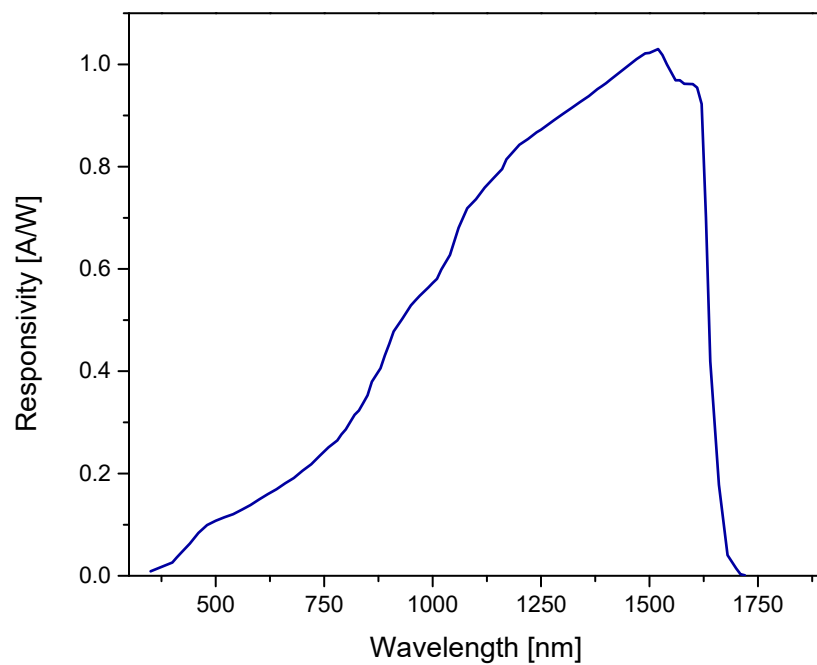
equation S4 can be simplified to:

$$\begin{aligned}
\frac{dN_c}{dz} &= \frac{K \sqrt{E_p}}{n_1 n_2 \tau \pi \omega_0^2} \frac{N_1 N_2}{E_g^2} F_{NTA}^{tot} \\
F_{NTA}^{tot} &= F_{NTA}^{tot} = x_2 F_{NTA}(x_1, x_2) + x_1 F_{NTA}(x_2, x_1) = \frac{(x_1 + x_2 - 1)^{3/2}}{2^6 x_1^3 x_2^3} (x_1 + x_2)^2
\end{aligned}
\tag{S6}$$

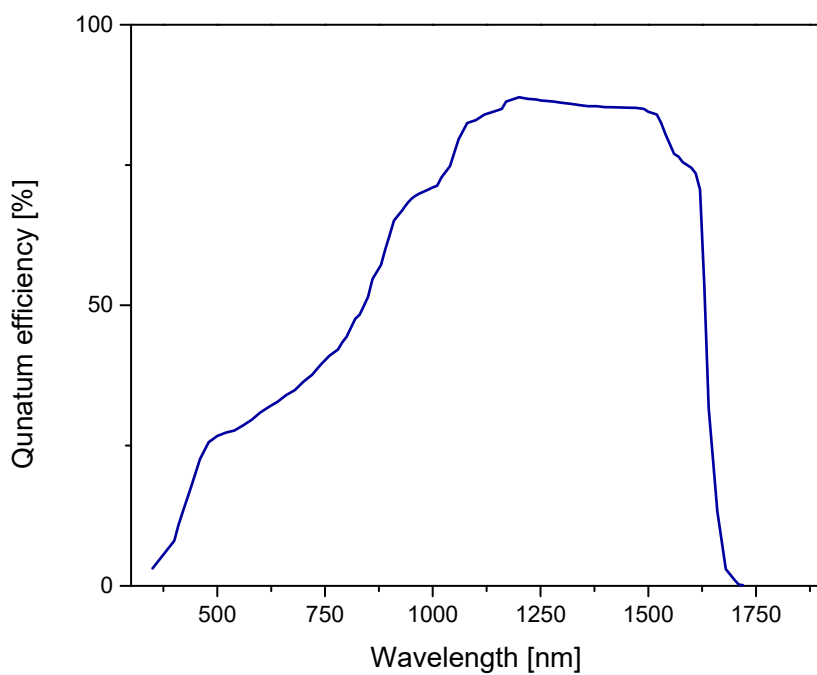
It is important to note that the photon, bandgap and Kane energies in equation (S6) are expressed in SI units, i.e. Joules. However, it is common to express energies in units of electron-Volts, which also define the typical units used for parameter K [1-3]. If eV is chosen as the unit for energy, then equations (S4) and (S6) are modified by a conversion factor $C = \frac{1}{1.6 \cdot 10^{-19}} \frac{eV}{J}$.

Supplementary Table 1. Experimental parameters.

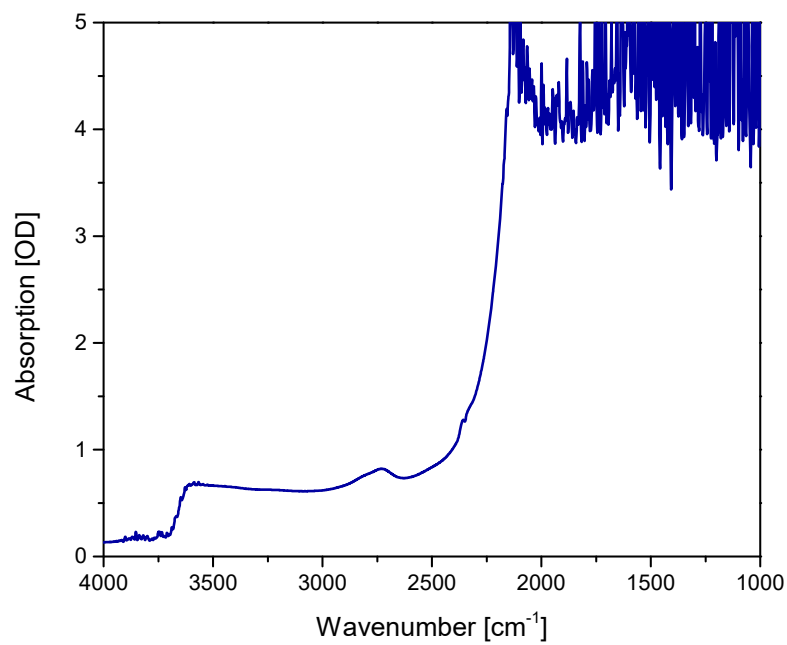
MIR pulse width	~110 fs
MIR power	130 μ W @ 1 kHz (accounted for camera window absorption)
NIR power	20 μ W @ 1 kHz
MIR spot size	3.5 mm
NIR spot size	4 mm
Pixel size	12 mm x 12 mm
CCD active area	1280x1024 pixels
Signal-to-noise ratio	
$10 \cdot \lg \left(\frac{S_{NTA}}{\sigma_{DTA}} \right)$	30 dB
Signal-to-noise ratio (mean power root)	
$20 \cdot \lg \left(\frac{S_{NTA}}{\sigma_{DTA}} \right)$	60 dB
Signal-to-background	
Signal-to-noise ratio	
$10 \cdot \lg \left(\frac{S_{NTA}}{S_{DTA}} \right)$	20 dB



Supplementary Figure S1. Spectral responsivity of the InGaAs camera.



Supplementary Figure S2. Spectrum of the quantum efficiency of the InGaAs camera.



Supplementary Figure S3. Absorption spectrum of the protective camera window (2 mm), which significantly attenuates the MIR radiation (OD=1.25 @ 4200 nm, OD>5 @ 5000 nm).

Supplementary Table 2. Relative comparison of experimental parameters used in the measurements with Si camera and InGaAs camera.

	Si	InGaAs
MIR power	1 mW @ 1 kHz (accounted for camera window absorption)	130 μ W @ 1 kHz (accounted for camera window absorption)
NIR power	120 μ W @ 1 kHz (accounted for camera window absorption)	20 μ W @ 1 kHz (accounted for camera window absorption)
Exposure time	100 ms (100 pulses @ 1 kHz)	60 μ s (1 pulse @ 1 kHz)
Quantum efficiency @ $\hbar\omega_{MIR} + \hbar\omega_{NIR}$	1%	80%

References

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