Single-Photon Software Lock-in for PL

Pipelines, Results, and Applications

Summary

1. Built two photon-counting lock-in pipelines:
   - Jakob et al. + Liu et al., yields a photon count.
   - Jakob et al. + Braun & Libchaber, yields modulation depth M and phase ϕ.

2. Explored different experimental conditions, each time comparing the lock-in and regular counting.
3. Implemented FDLM : extracting τ from $\phi(\omega)$ and/or $M(\omega)$.

Deliverables: CLI tools (reader, lock-ins), simple decision rules for when to use lock-in vs counting, and a (preliminary) FDLM workflow.

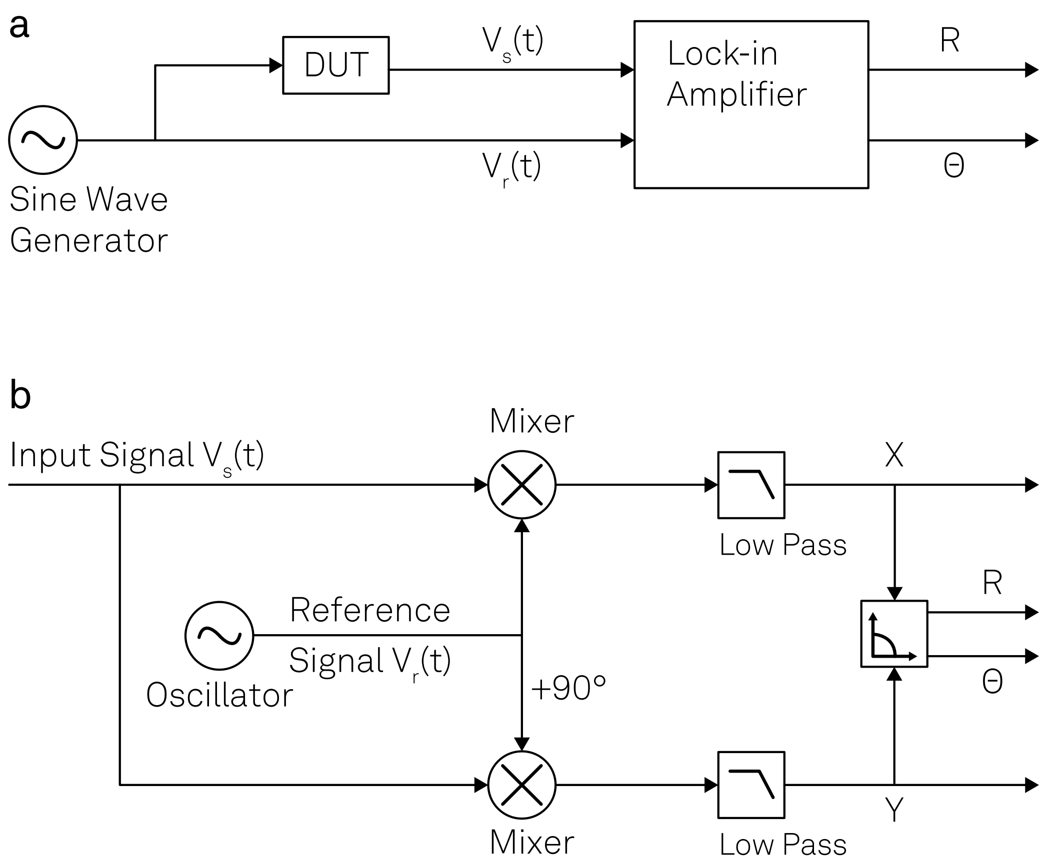
Quick Background

Classical:

- Inputs: Signal V_s(t), Reference V_r(t)
- Mixing (multiplication) -> low pass filtering (integration)
- Outputs: R, theta

Photon counting:

- Inputs: {t_p}, {t_m} taken in [0, T]
- Lock-in routine
- Outputs: Photon count (<= card({t_p})), mod. depth, theta

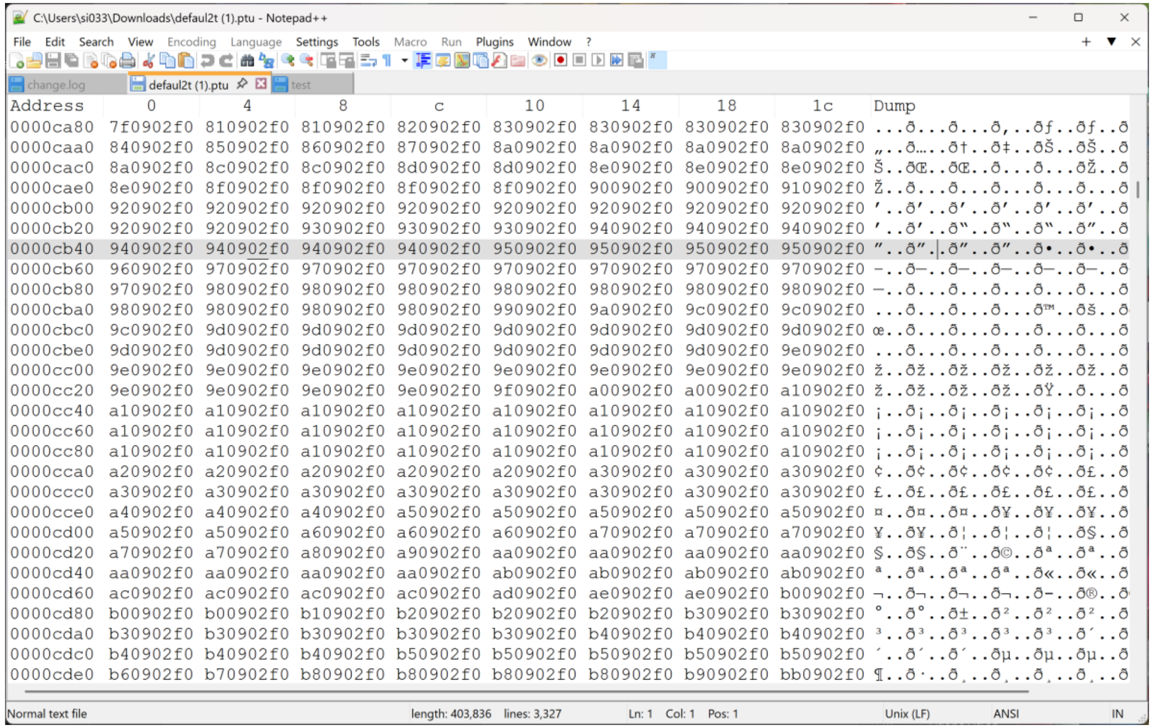


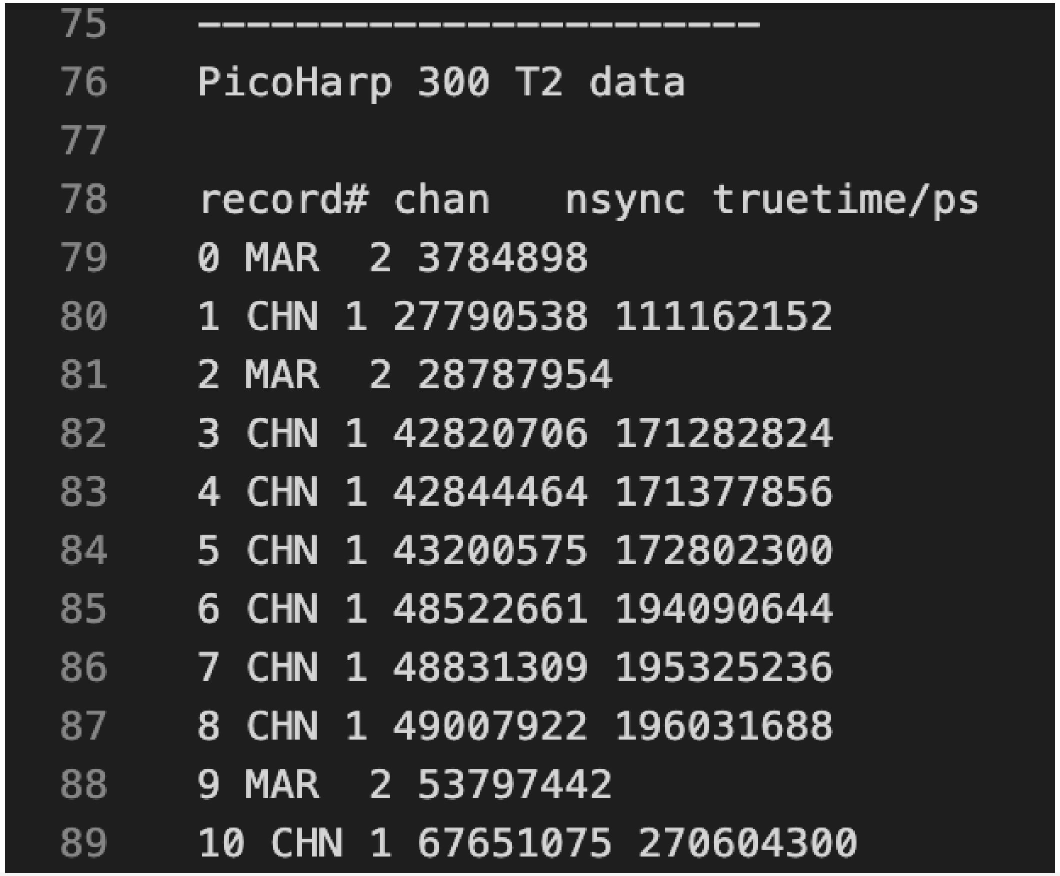
Physical lock-in inner workings

Hardware & Raw Data

Setup: Laser → Chopper (out: TTL) → Optics → SPAD (out: NIM) → PicoHarp-330 (T2).

Raw data (.PTU) (look inside, bits) -> decode -> .txt
In practice: .PTU -> lock-in (.txt files are huge)

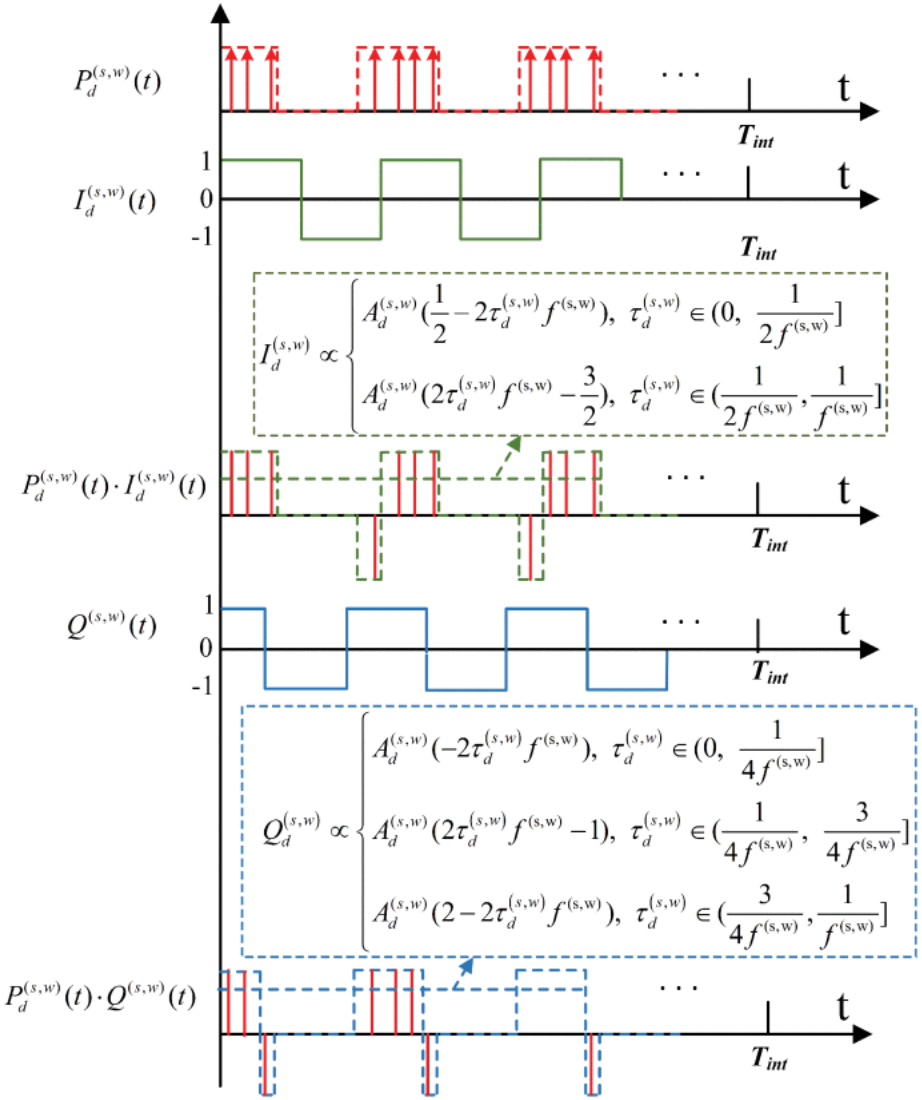




Algorithms

Liu et al. (reference-weighted counting) and Braun & Libchaber

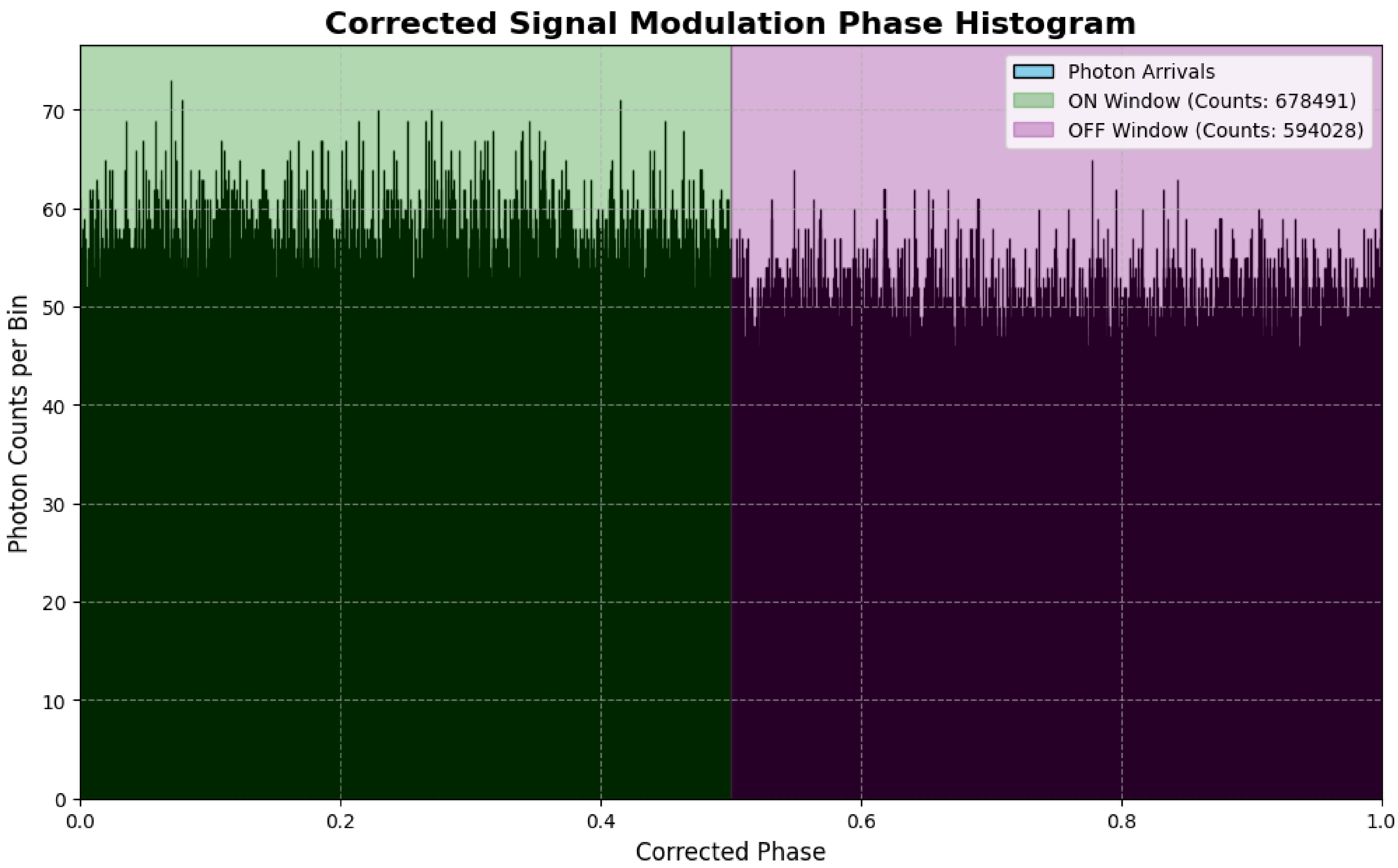
Algorithm A — Liu (reference-weighted counting)



- Compute instantaneous phase for each photon from reference markers.

- Accumulate I = Σ square(ϕ), Q = Σ square(ϕ + 90°) over windows
- Signal Strength (or Photon Count) = norm(I + iQ), Phase = atan2(Q, I)

- Jakob Augmentation: dynamic references, better per-photon phase calculation.
- Main use cases: Multiple sources and responses, daylight operations.



Corrected Signal Modulation Phase Histogram
Photon Arrivals
ON Window (Counts: 678491)
OFF Window (Counts: 594028)
Photon Counts per Bin
Corrected Phase
0
10
20
30
40
50
60
70
0.0
0.2
0.4
0.6
0.8
1.0

Algorithm A — Braun & Libchaber

Algorithm 1 Photon-Counting Lock-in Detection

function PhotonCountingLockIn

  // Step 1: Record photon counts in time bins
  $N_{raw} \leftarrow$ empty array
  for each time bin t_n with interval $1/f_{sample}$ **do**
    $N_{raw}[n] \leftarrow$ number of photons detected in bin
  end for

  // Step 2: Create functional basis
  $R_{raw\_avg} \leftarrow \frac{1}{T}\sum_{n=0}^{T-1} R_{raw}(t_n)$ ▷ Time average
  Find zero crossings t_z with positive slope
  $f_{ref}, \phi_{ref} \leftarrow \text{LinearRegression}(t_z)$ ▷ Extract frequency and phase
  $R_X(t_n) \leftarrow \sin(2\pi f_{ref} t_n + \phi_{ref})$ ▷ Sine reference
  $R_Y(t_n) \leftarrow \cos(2\pi f_{ref} t_n + \phi_{ref})$ ▷ Cosine reference

  // Step 3: Project counts onto basis
  $I \leftarrow \frac{1}{T}\sum_{n=0}^{T-1} N_{raw}(t_n)$ ▷ Average count rate
  $N(t_n) \leftarrow N_{raw}(t_n) - I$ ▷ Center around zero
  $I_X \leftarrow \frac{\sum_n N(t_n)\cdot R_X(t_n)}{\sqrt{\sum_n R_X(t_n)^2}}$ ▷ In-phase component
  $I_Y \leftarrow \frac{\sum_n N(t_n)\cdot R_Y(t_n)}{\sqrt{\sum_n R_Y(t_n)^2}}$ ▷ Quadrature component

  // Step 4: Calculate amplitude and phase
  $A \leftarrow \frac{\sqrt{I_X^2 + I_Y^2}}{I}$ ▷ Relative amplitude
  $\phi \leftarrow \arctan \frac{I_Y}{I_X}$ ▷ Signal phase
  return A, ϕ, I

end function

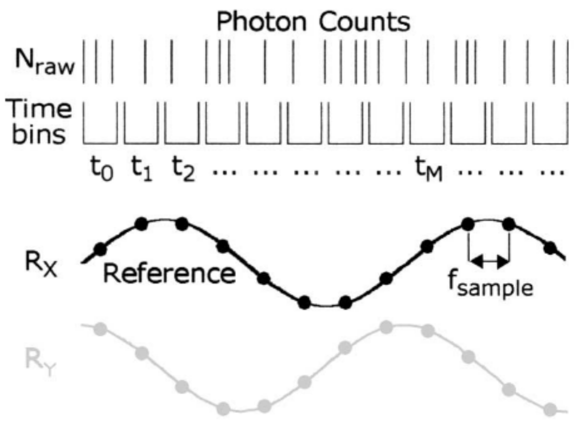


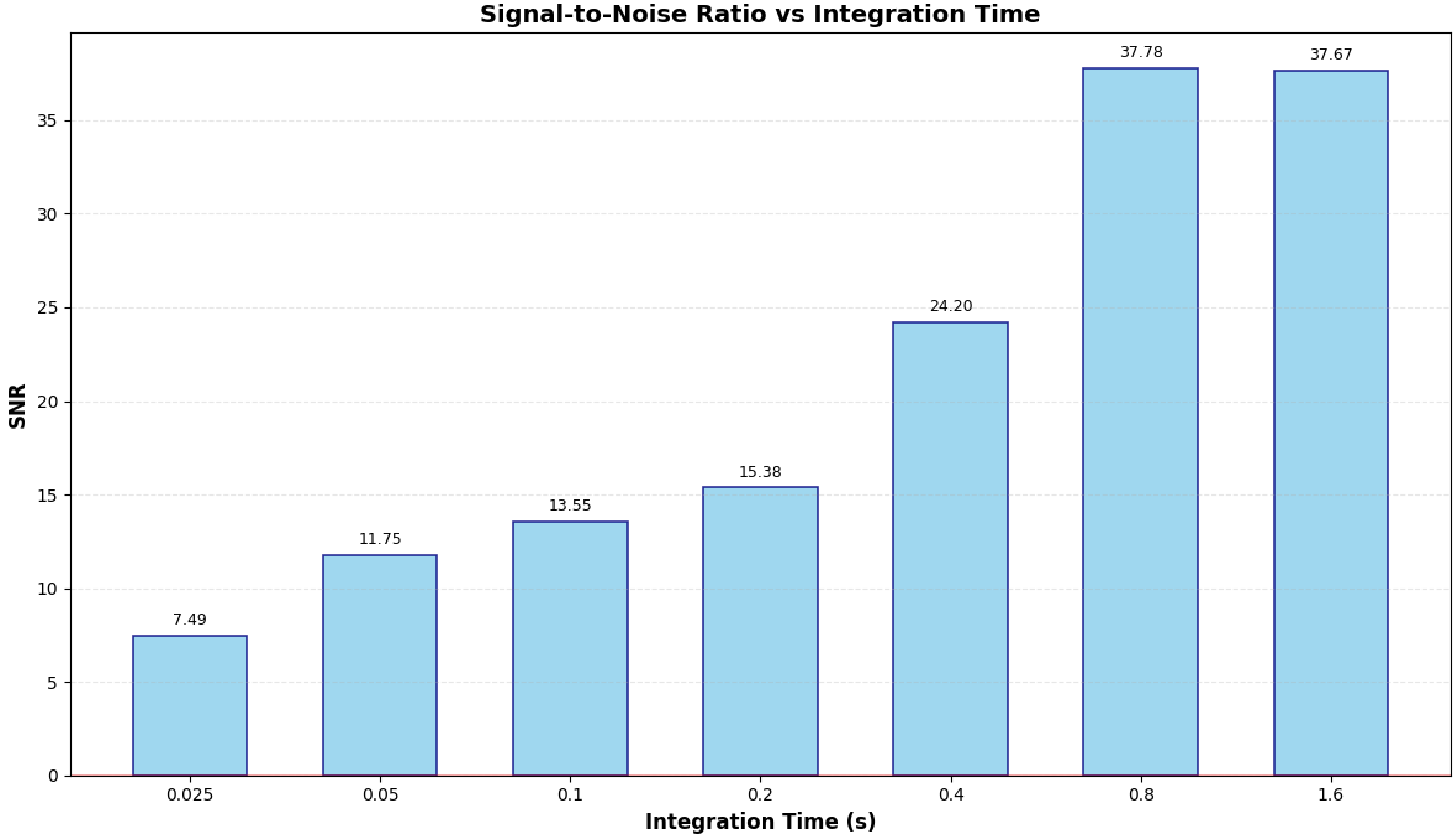
Fig. 1. Implementation of the photon-counting lock-in. Projections of time-binned counts N_{raw} to sinusoidal reference R_X and cosinusoidal reference R_Y are time averaged to yield the small I_X and I_Y signals of the lock-in.

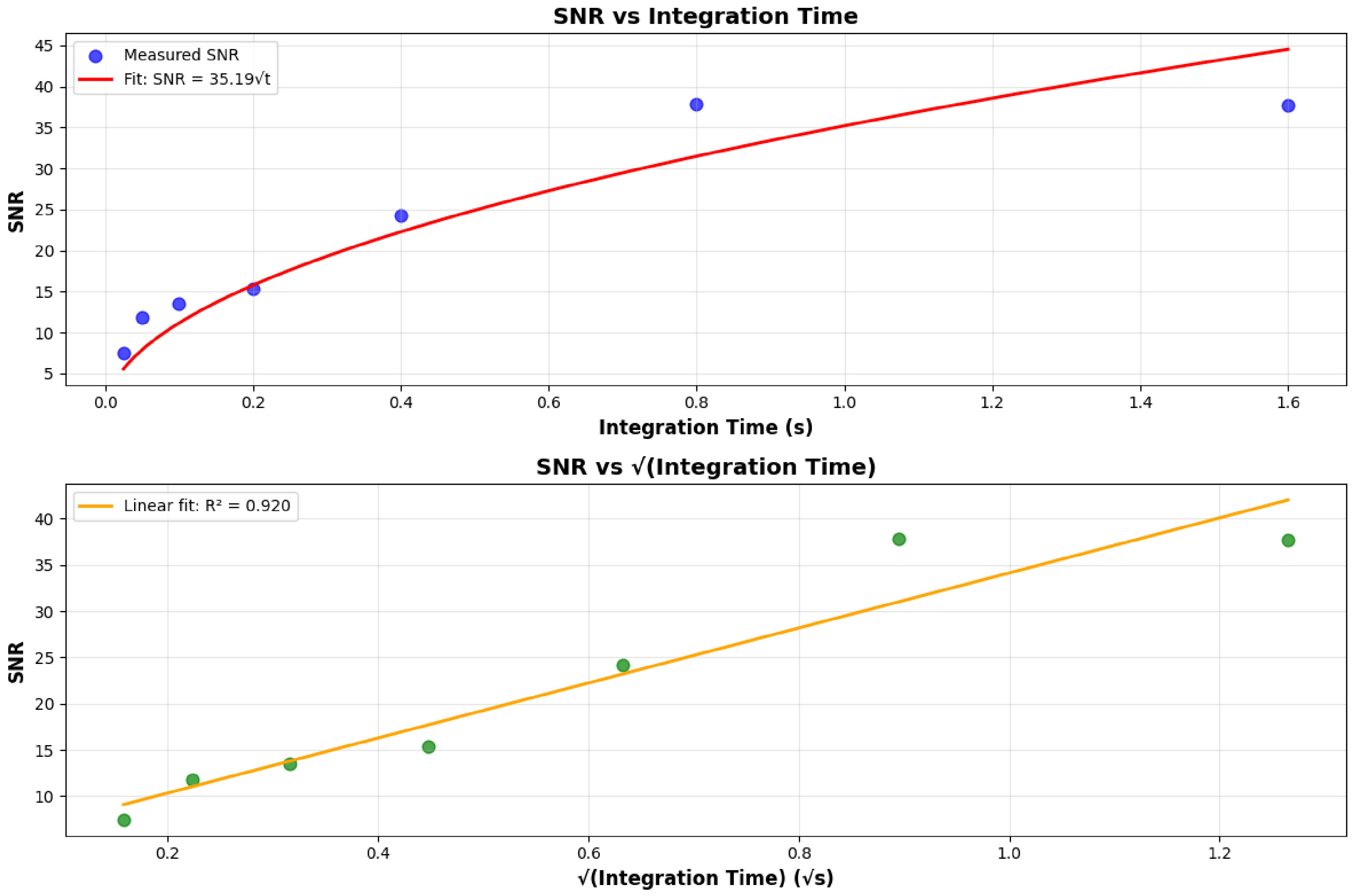
Jakob et al. Augmentation: no need to bin, we resolve each individual photon arrival.

Baselines & Statistics

OFF estimates background; subtract from ON.

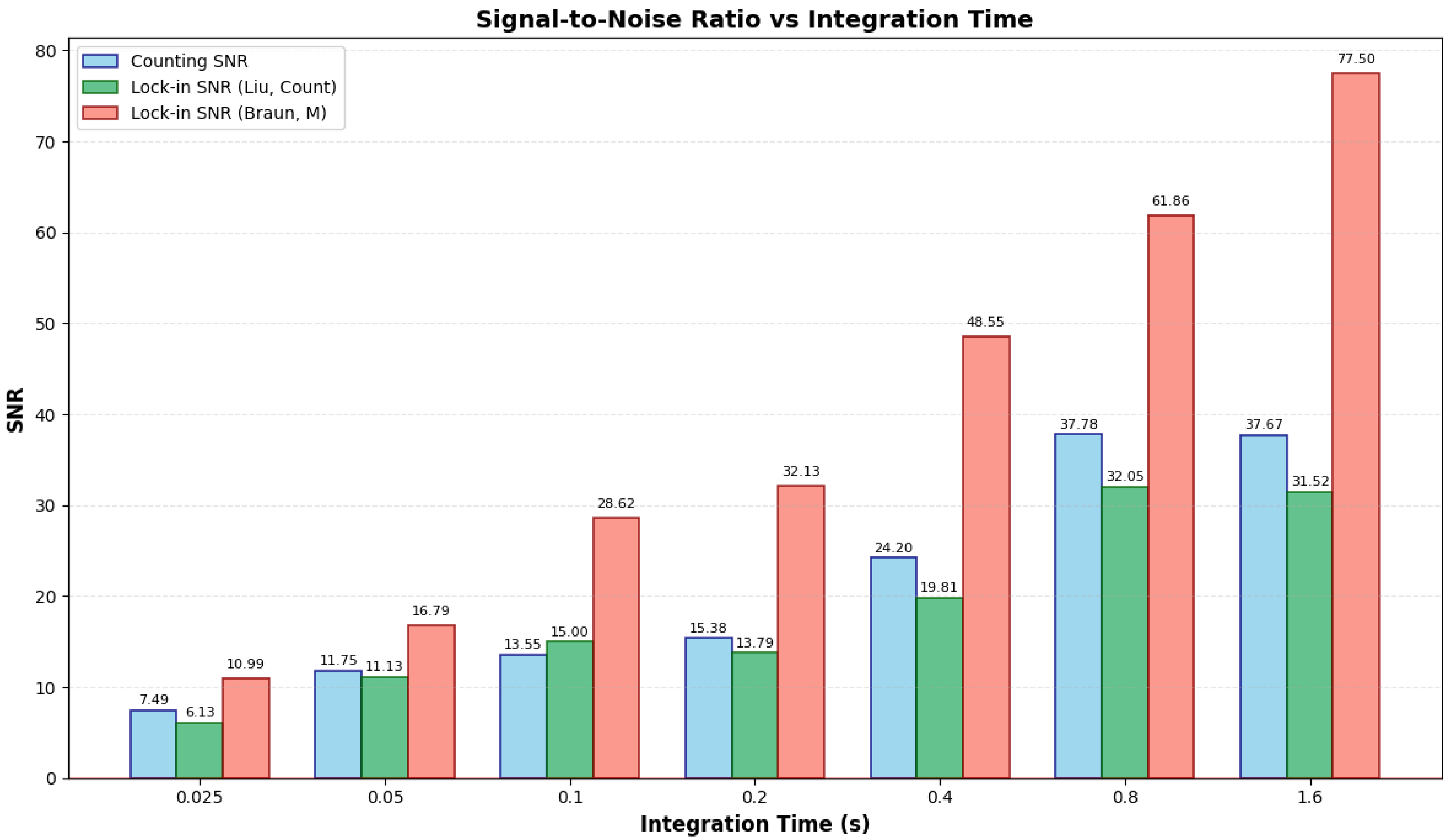
SNR = mean/std

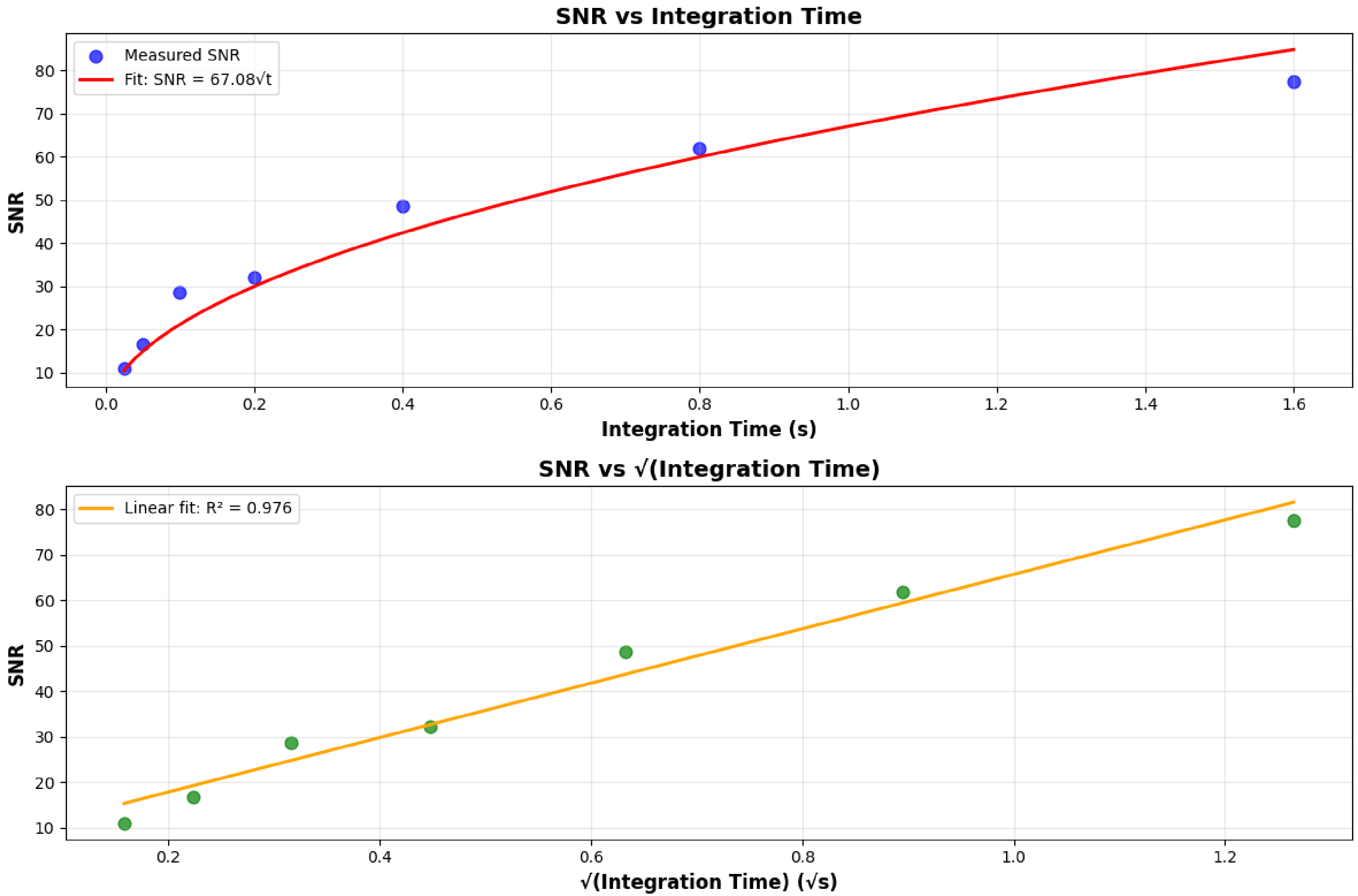



SNR vs Integration Time
Measured SNR
Fit: SNR = 35.19√t
SNR
Integration Time (s)
SNR vs √(Integration Time)
Linear fit: R² = 0.920
SNR
√(Integration Time) (√s)

SNR Results

SNR — Lock-in vs ON/OFF



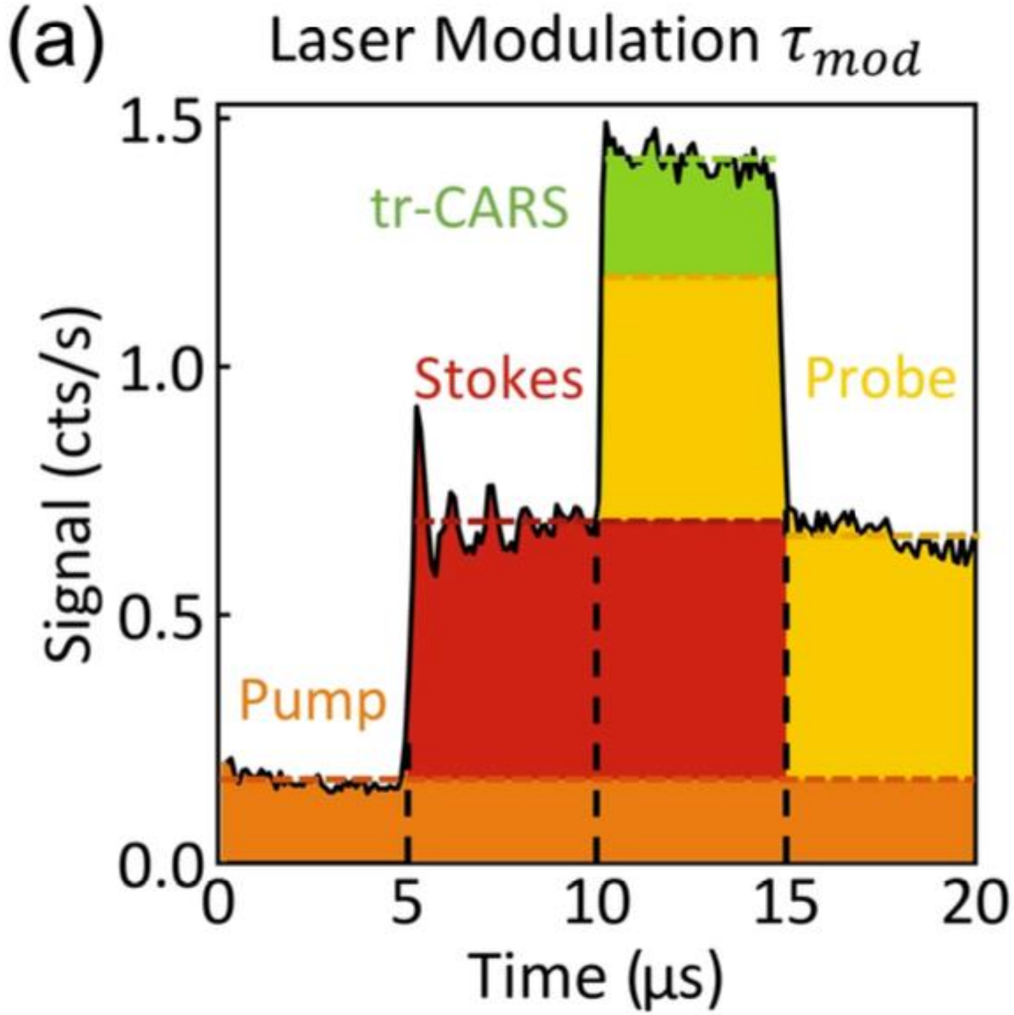


SNR vs Integration Time
Measured SNR
Fit: SNR = 67.08√t
SNR
Integration Time (s)
SNR vs √(Integration Time)
Linear fit: R² = 0.976
SNR
√(Integration Time) (√s)

In photon counting without locking, we can measure only photon counts I at the noise limit [Eq. (4)]. When we use the photon-counting lock-in, we can also detect the small-signal amplitude and small-signal phase at the same fundamental detection limit [Eqs. (5)] as shown in Fig. 3.

Some Applications

Multiplexing, Lifetimes from Phase and Modulation



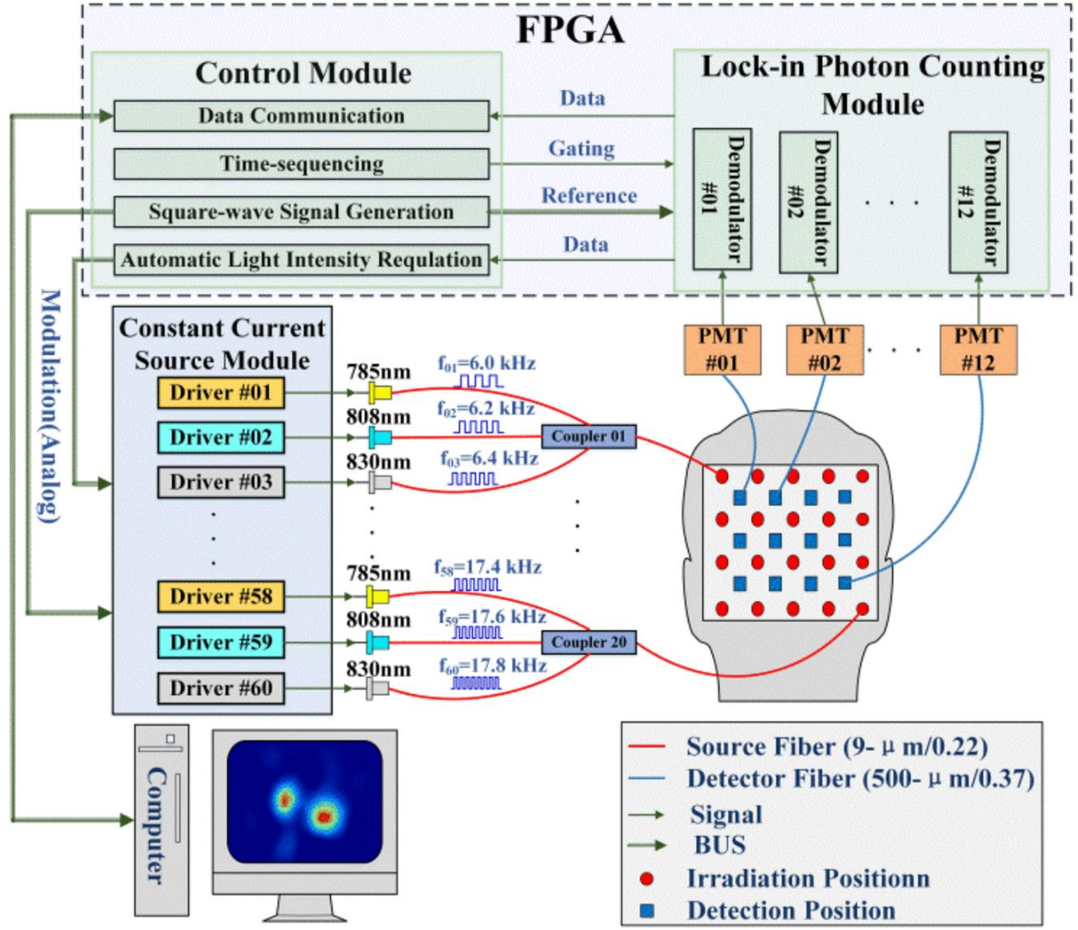
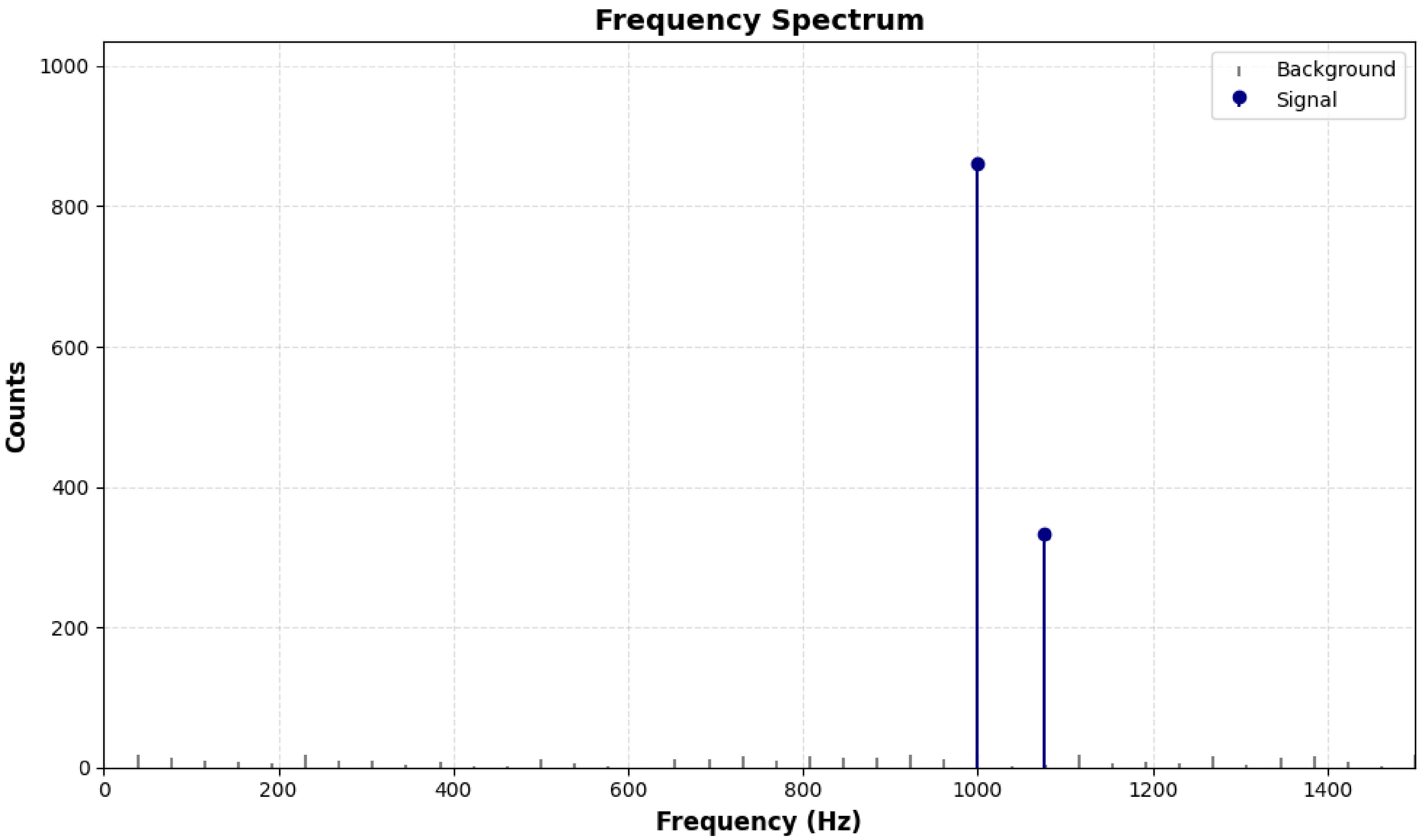
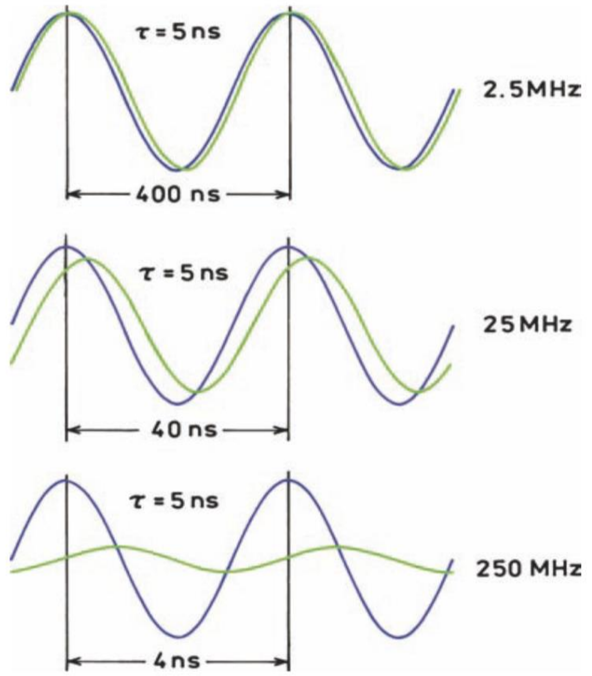


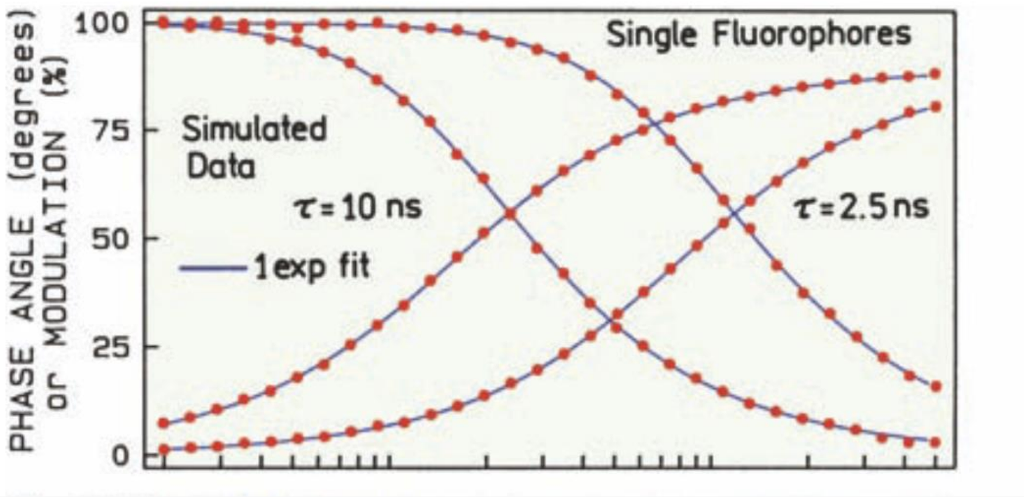
FIGURE 1. Schematic of the fNIRS-DOT instrument.



Frequency Spectrum
Background
Signal
Counts
Frequency (Hz)
0
200
400
600
800
1000
1200
1400

FDLM Theory (single-exponential)





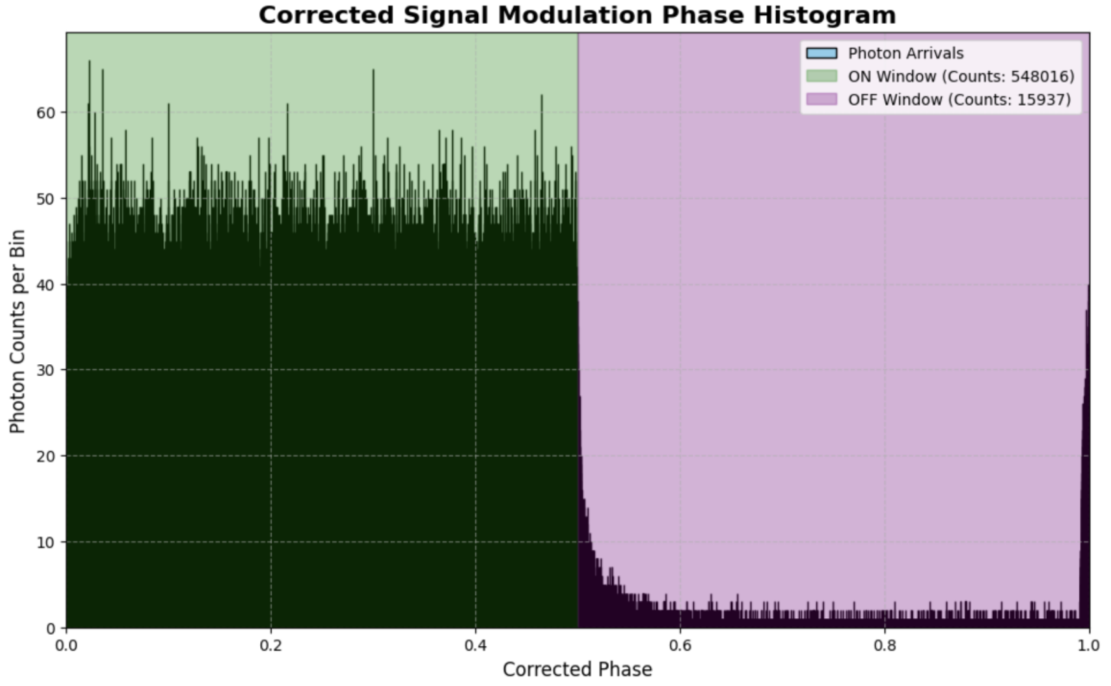
As modulation frequency is increased:

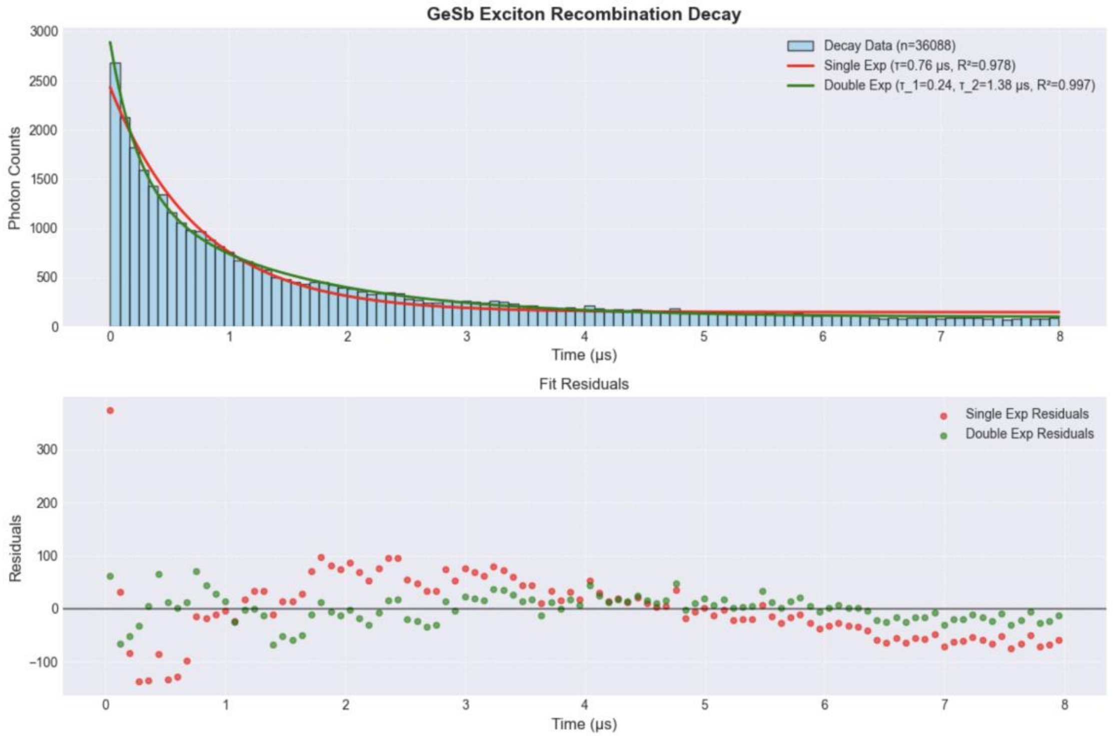
- Modulation decreases from 1 to 0
- Phase increases from 0, pi/2

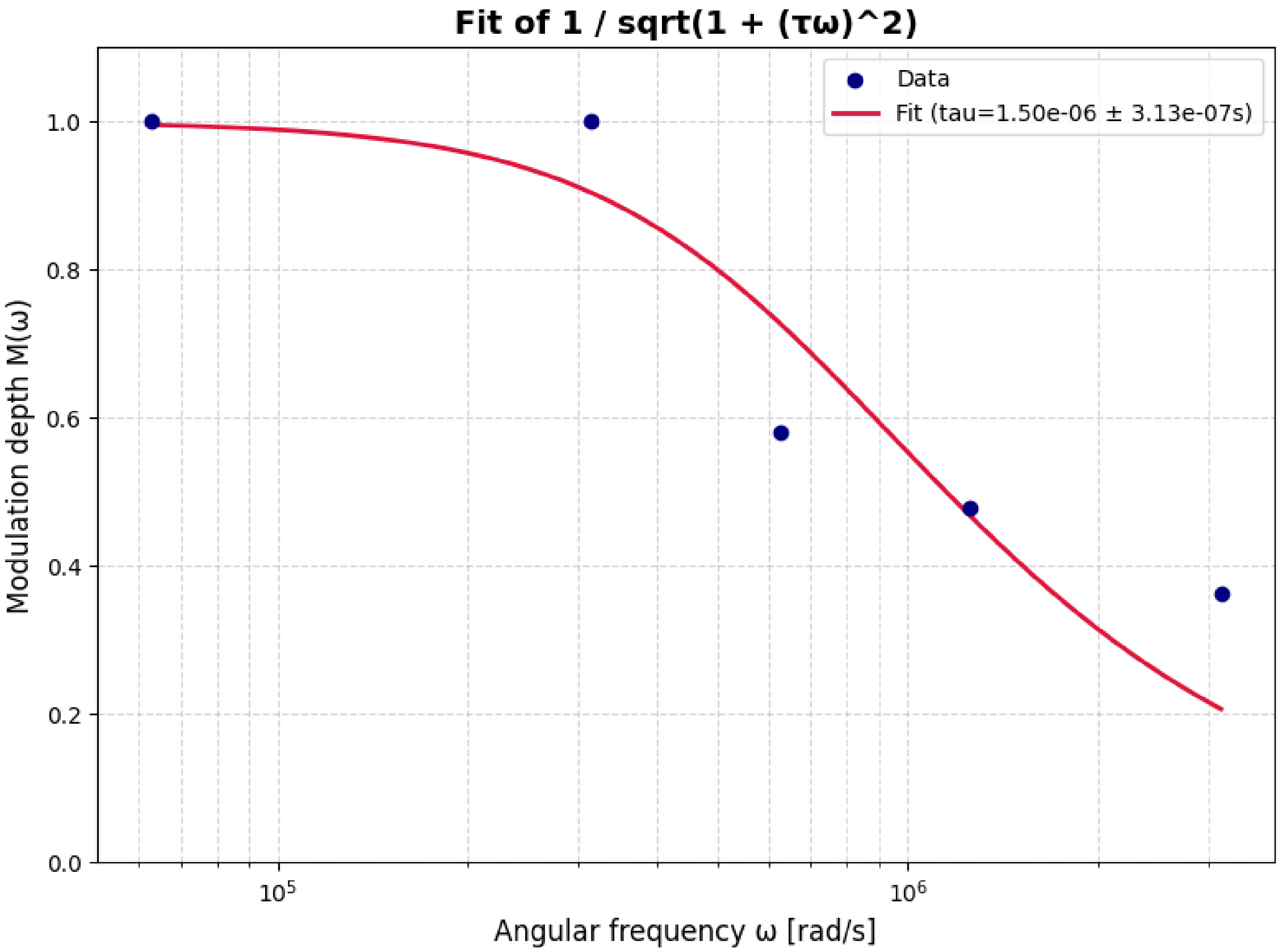
Idea: Extract lifetime from dynamics M(ω) and ϕ(ω). Models are especially simple if we assume single exponential decay:

- $M(\omega) = 1 / \sqrt{(1 + (\omega\tau)^2)}$
- $\tan \phi(\omega) = \omega \tau$

Use phase and/or modulation to fit τ; amplitude a bit more straightforward.


Corrected Signal Modulation Phase Histogram
Photon Arrivals
ON Window (Counts: 548016)
OFF Window (Counts: 15937)
Photon Counts per Bin
Corrected Phase


GeSb Exciton Recombination Decay
Decay Data (n=36088)
Single Exp (τ=0.76 μs, R²=0.978)
Double Exp (τ_1=0.24, τ_2=1.38 μs, R²=0.997)
Photon Counts
Time (μs)
Fit Residuals
Single Exp Residuals
Double Exp Residuals
Residuals
Time (μs)



Fit of 1 / sqrt(1 + (τω)^2)
Data
Fit (tau=1.50e-06 ± 3.13e-07s)
Modulation depth M(ω)
1.0
0.8
0.6
0.4
0.2
0.0
10^5
10^6
Angular frequency ω [rad/s]

FDLM Pipeline

1) Frequency sweep: collect TTTR T2 at $f_1 \ldots f_n$.
2) Braun(f) → (M, ϕ).
3) Fit τ from M(ω).

```
└─[$] <git:(main*)> python3 braun_lockin_original.py data/GeSb/10mW_GeSb_10s_10KHz_50pctdutycycle_000.ptu
File: data/GeSb/10mW_GeSb_10s_10KHz_50pctdutycycle_000.ptu
RESULT — Braun photon-counting lock-in:
  f_ref              : 9999.860445 Hz
  f_sample           : 499993.022250 Hz   (M = 50 bins/period)
  bins / photons     : 4999797 / 563984
  I_dc (mean/bin)    : 0.112801 counts/bin
  A (mod depth)      : 1.21884303 (dimensionless)
  phi                : 3.118130083 rad  (178.655694 deg)
  bins_per_period    : 50
  trimmed periods    : 1 at start and end
```

Lessons Learned, Recs, If I had more time...

- Lock-in is not a free lunch: with stable background, ON–OFF sets a high bar in terms of SNR.
- Lock-in enables measurements beyond frequency counting: modulation, phase, multiple responses, daylight measurements (!)

- Use lock-in when we know of periodic noise, nearby in the band.
- When we need to enable measurements stated above.
- Otherwise, not worth the overhead. Introduces extra time and complexity (sources of error).

- Multi-exponential FDL fits, and further improvements to the models (regularization, error propagation, etc.)
- Cross-validation against TCSPC decay fits
- Dual-modulation experiment ?!

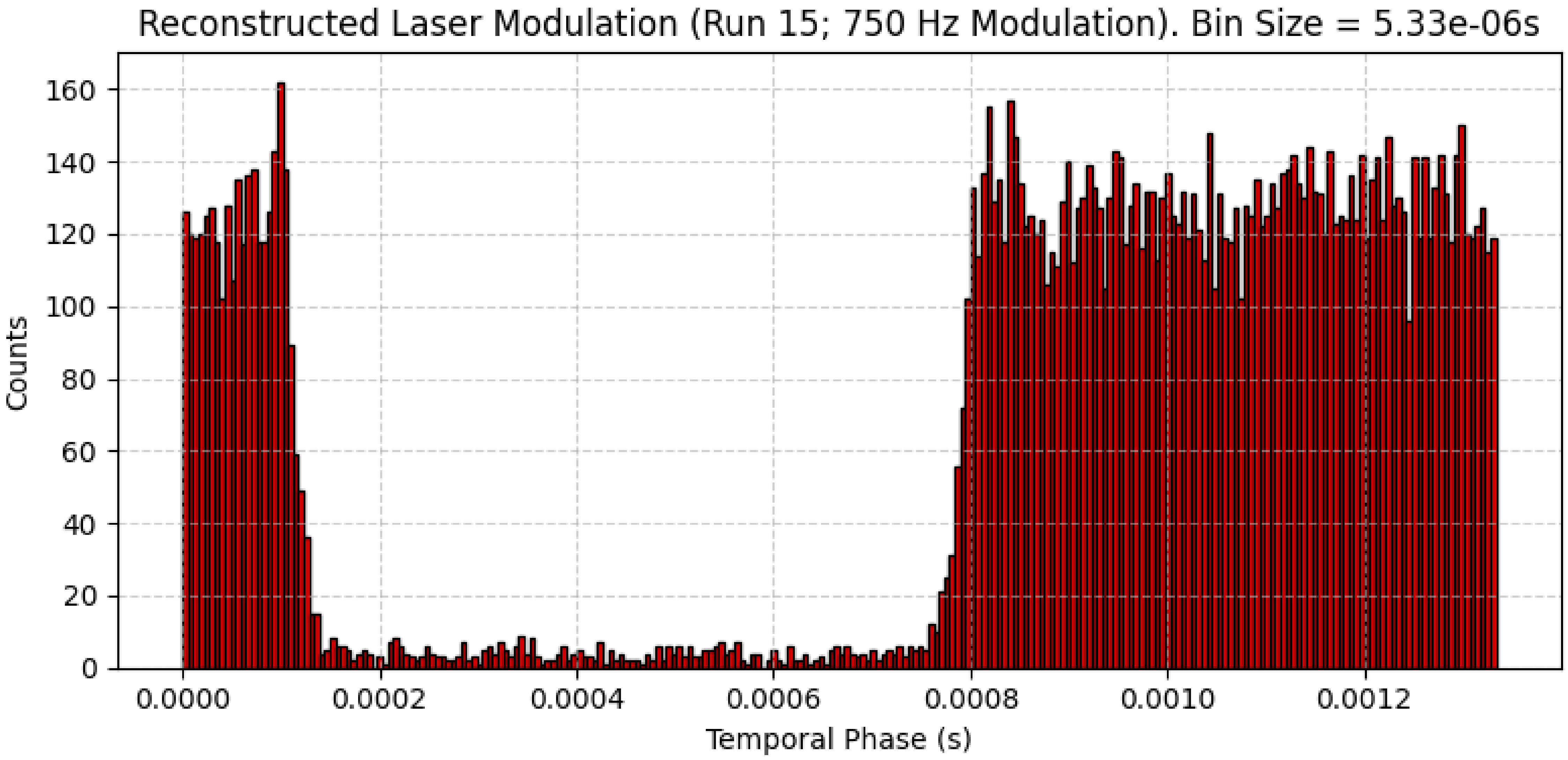
- Jakob, L. A., Deacon, W. M., Hicks, O., Manyakin, I., Ojambati, O. S., Traxler, M., & Baumberg, J. J. (2021). *Single-photon multiclock lock-in detection by picosecond timestamping*. **Optica**, 8(12), 1646–1653. https://doi.org/10.1364/OPTICA.441487

- Braun, D., & Libchaber, A. (2002). *Computer-based photon-counting lock-in for phase detection at the shot-noise limit*. **Optics Letters**, 27(16), 1418–1420. https://doi.org/10.1364/OL.27.001418

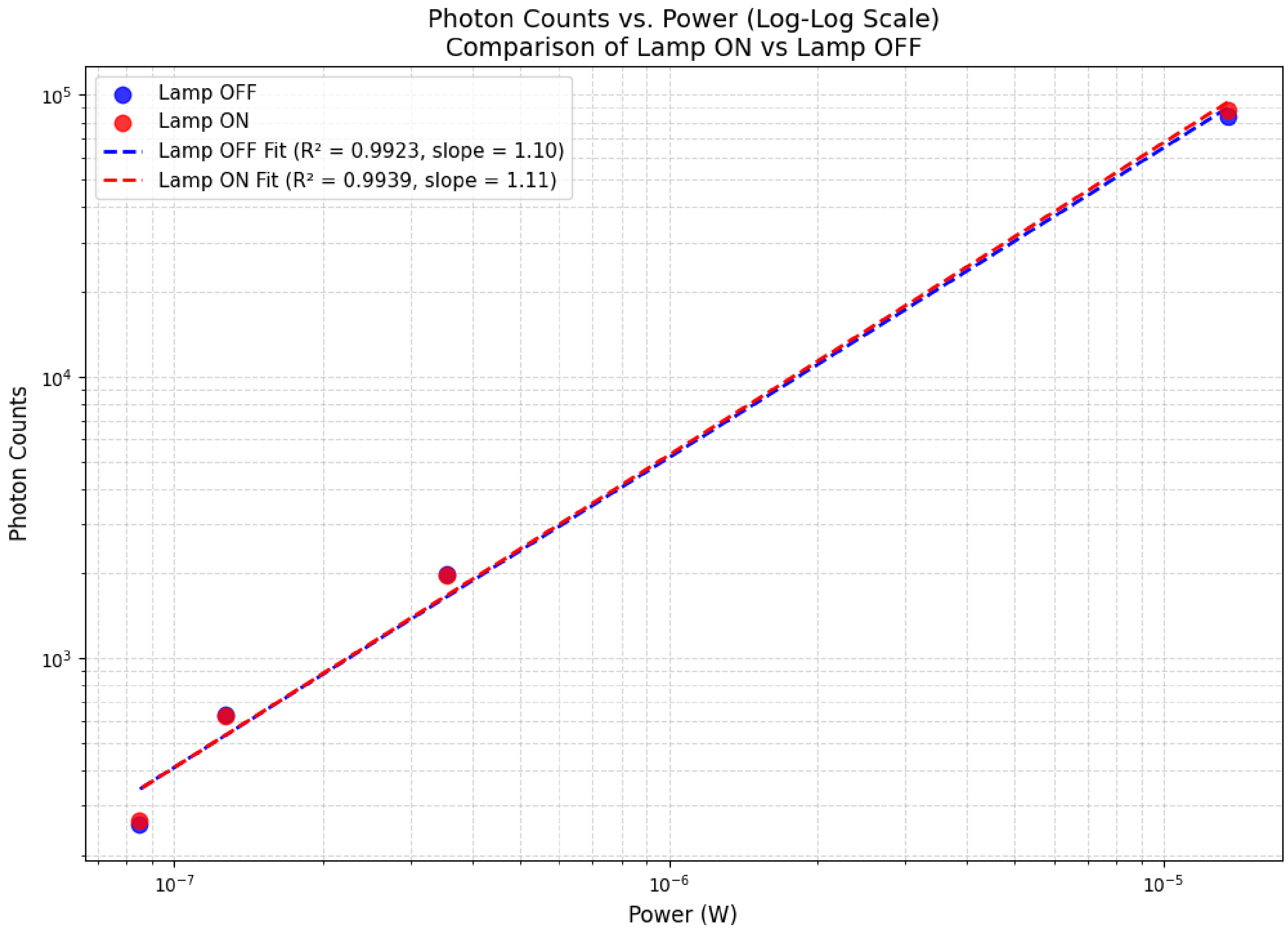
- Liu, D., Wang, B., Pan, T., Li, J., Qin, Z., Zhang, L., Zhou, Z., & Gao, F. (2015). *Toward quantitative near infrared brain functional imaging: Lock-in photon counting instrumentation combined with tomographic reconstruction*. **IEEE Transactions on Biomedical Engineering**, 62(12), 2941–2951. https://doi.org/10.1109/TBME.2015.2450217

- PicoQuant GmbH. (2013). *PicoHarp 300: Picosecond histogram accumulating real-time processor — User's manual and technical data* (Software version 2.3). Berlin, Germany: PicoQuant GmbH.

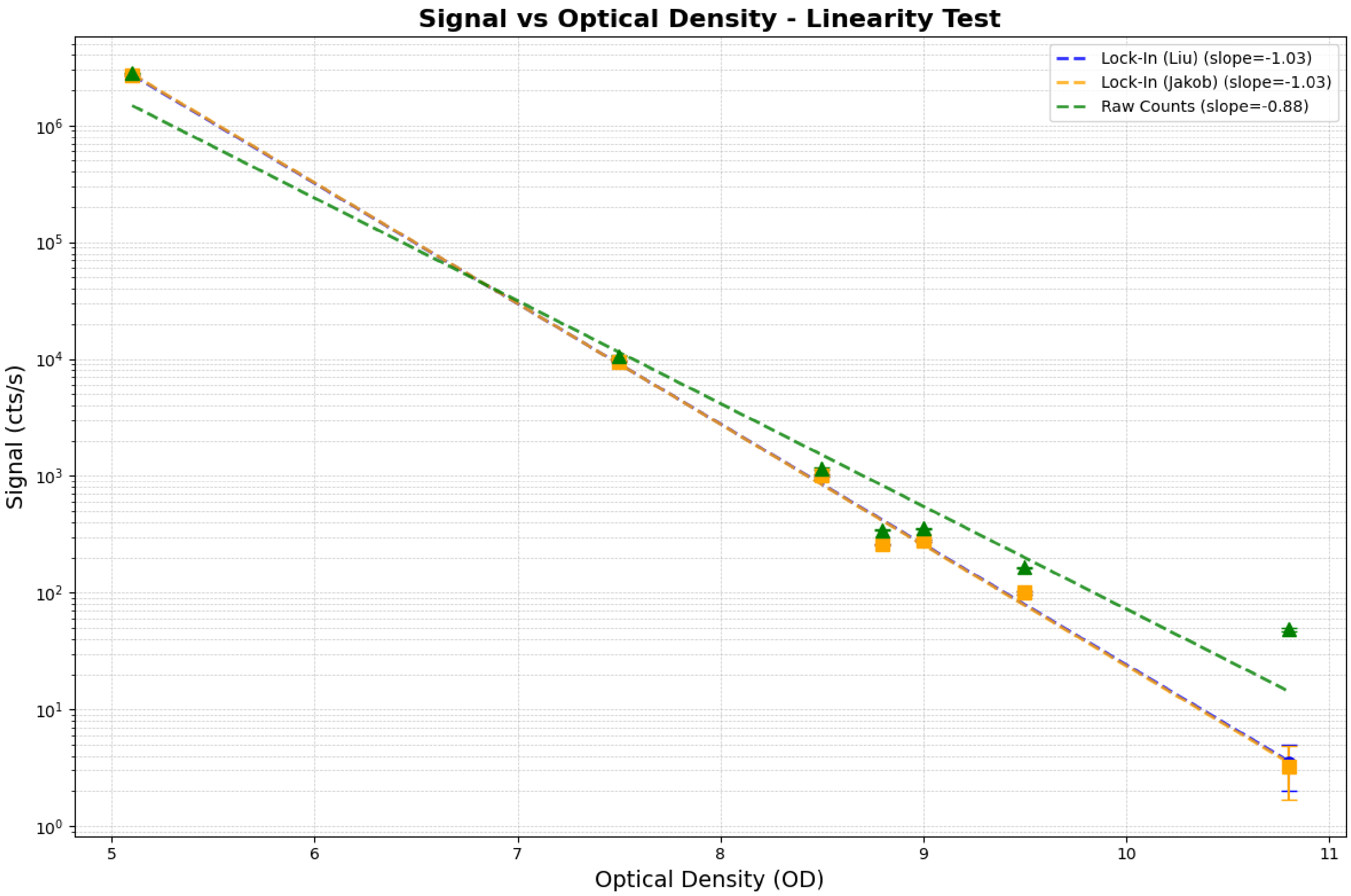
- Lakowicz, J. R. (2006). *Principles of Fluorescence Spectroscopy* (3rd ed.). Springer. (Frequency-Domain Lifetime Measurements, Ch. 5).



Typical Reconstructred ON/OFF Run (PicoHarp 300 data)



Lock-In Outputs at Different Laser Powers, High and Low Background.



Signal vs Optical Density - Linearity Test
Lock-In (Liu) (slope=-1.03)
Lock-In (Jakob) (slope=-1.03)
Raw Counts (slope=-0.88)
Signal (cts/s)
Optical Density (OD)
10^0
10^1
10^2
10^3
10^4
10^5
10^6
5
6
7
8
9
10
11