

# Absolute fluorescence and absorption measurements over a dynamic range of $10^6$ with cavity-enhanced laser-induced fluorescence

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## Metadata supporting open data access

This document sets out the metadata of the original data recorded as published in this paper in support of the open data access.

### Figure 2: LIF and CRD transients

File: Fig02ab TransOn\_ResBPPEB.dat

LIF and CRD transients recorded on resonance (vibronic transition of BPPEB at 319.44 nm).

Molecular-beam pulse optimised to temporally overlap with the laser.

Column 1: time after Nd:YAG Q-switch trigger (in  $\mu$ s)

Column 2: signal of ring-down PMT (in mV)

Column 3: signal of LIF PMT (in mV)

File: Fig02cd TransOffResBPPEB.dat

LIF and CRD transients recorded off resonance (BPPEB at 321 nm).

Molecular-beam pulse optimised to temporally overlap with the laser.

Column 1: time after Nd:YAG Q-switch trigger (in  $\mu$ s)

Column 2: signal of ring-down PMT (in mV)

Column 3: signal of LIF PMT (in mV)

### Figure 3a: LIF signal dependence on incident laser intensity

File: Fig03a 0-1mbar.dat

Cavity filled with 0.1 mbar acetone.

Each data point corresponds to 5000 laser shots.

Column 1: averaged time-integrated ring-down transient (arb. units)

Column 2: standard deviation

Column 3: standard error

Column 4: averaged time-integrated LIF transient (arb. units)

Column 5: standard deviation

Column 6: standard error

File: Fig03a 0-3mbar.dat

Cavity filled with 0.3 mbar acetone.

Each data point corresponds to 5000 laser shots.

Columns identical to the 0.1 mbar file above.

### Figure 3b: Fractional absorptions from CRDS and CELIF measurements

File: Fig03b N2-Rayleigh.csv

Col 1: pressure [mbar], reading from calibrated baratron  
Col 2: [blank]  
Col 3: CELIF signal (fraction of time-integrated LIF transient over time-integrated CRD transient)  
Col 4: [blank]  
Col 5: ring-down time [ $\mu$ s]  
Col 6: [blank]  
Col 7:  $\alpha_{\text{CRD}}$  [ $\text{cm}^{-1}$ ]  
Col 8: [blank]  
Col 9: time-integrated LIF signal [arb. units]  
Col 10: time-integrated CRD signal [arb. units]  
Col 11: CELIF signal with background subtraction  
Col 12:  $\alpha_{\text{CELIF}}$  [ $\text{cm}^{-1}$ ]  
Cols 13,14: [unused]

The data analysis to produce Fig. 3(b) only uses Columns 1, 3 and 5 although Column 3 is generated from the entries reported in Columns 9 and 10.

### Figure 4: BPEB spectra via CRDS and CELIF

File: Fig04ab Signal7.txt

BPEB spectrum with high density of BPEB, hot nozzle (135 °C), optimal temporal overlap of molecular beam pulse and laser.

[Ignore the description of column in the file.]

Col 1: wavelength [nm]  
Col 2: ring-down time [s] (averaged over 10 laser shots per wavelength)  
Col 3: standard deviation of ring-down time  
Col 4: initial intensity of ring-down fitted decay [V] (average)  
Col 5: standard deviation of initial intensity  
Col 6: time-integrated ring-down transient [arb. units] (average)  
Col 7: standard deviation of Col 6  
Col 8: time-integrated LIF transient [arb. units] (average)  
Col 9: standard deviation of Col 8  
Col 10: CELIF signal (from Col 8 over Col 6)

File: Fig04ab Blank7.txt

Background spectrum: nozzle trigger set such that the molecular beam pulse appears after the laser pulse.

Columns as above.

File: Fig04cd Signal5.txt

BPEB spectrum with medium density of BPEB, nozzle temperature (115 °C), optimal temporal overlap of molecular beam pulse and laser.

Columns as above.

File: Fig04cd Blank5.txt

Background spectrum: nozzle trigger set such that the molecular beam pulse appears after the laser pulse.

Columns as above.

File: Fig04ef Signal1.txt

BPEB spectrum with low density of BPEB, nozzle temperature (75 °C), optimal temporal overlap of molecular beam pulse and laser.

Columns as above.

File: Fig04ef Blank1.txt

Background spectrum: nozzle trigger set such that the molecular beam pulse appears after the laser pulse.

Columns as above.

### Figure 5: Limits of detection

A data set of 40 files contributes to this figure. The filenames are coded as follows:

Fig05 *ttt\_sr\_vvv.txt*

*ttt*: nozzle temperature [ °C], increasing temperatures lead to higher BPEB concentrations.

*s*: S nozzle timing optimised to best temporal overlap with laser – signal measurement;  
B nozzle mistimed, molecular beam pulse after laser pulse – background measurement.

*r*: 1 means on resonance (319.69 nm) – signal measurement;  
0 means off resonance (320.98 nm) – background measurement.

*vvv*: Voltage of LIF PMT [V].

The data was recorded using increasing nozzle temperatures leading to higher BPEB densities in the molecular beam. The LIF PMT gain needed to be lowered with increasing signal level. In order to ensure a correct internal calibration, an on-resonance signal and a background spectrum were recorded with a high and low PMT voltage for the same BPEB density (nozzle temperature).

Format of the above files:

Col 1: time-integrated LIF transient [arb. units]

Col 2: time-integrated ring-down transient [arb. units]

Col 3: resulting CELIF signal

Cols 4–6: Same as Cols 1–3, but using a different time interval for the integration; not used in the analysis.

Col 7: fitted ring-down time [μs]

Col 8: mean-squared residual of the ring-down-time fit.

Cols 9–13: [unused, legacy in LabView program]

### Figure 6a: single-pass LIF spectrum

File: Fig06a sp-LIF.txt

BPEB spectrum with high density of BPEB, hot nozzle (135 °C), optimal temporal overlap of molecular beam pulse and laser.

[Ignore the units listed in the file.]

Col 1: wavelength [nm]

Col 2: LIF signal normalised by laser intensity [arb. units]

Col 3: raw signal from LIF PMT (time-integrated) [arb. units]

Col 4: [unused]

Col 5: laser intensity: signal from pyro-electric detector placed after the exit window [arb. units]

Col 6: background-subtracted and normalised LIF signal (max. = 1) [used for plotting]

Each data point is averaged over 10 laser shots.

### Figure 6b and 6c: LIF spectra with cavity

File: Fig06bc C-E-LIF.txt

BPEB spectrum with high density of BPEB, hot nozzle (135 °C), optimal temporal overlap of molecular beam pulse and laser.

[Ignore the units listed in the file.]

Col 1: wavelength [nm]

Col 2: ring-down time [s] (averaged over 10 laser shots per wavelength)

Col 3: standard deviation of ring-down time

Col 4: initial intensity of ring-down fitted decay [V] (average)

Col 5: standard deviation of initial intensity

Col 6: time-integrated ring-down transient [arb. units] (average)

Col 7: standard deviation of Col 6

Col 8: time-integrated LIF transient [arb. units] (average)

Col 9: standard deviation of Col 8

Col 10: CELIF signal,  $S^{\text{CELIF}}$ , (from Col 8 over Col 6)

Col 11: standard deviation of Col 10

Col 12:  $S^{\text{LIF}}$ , rescaled from Col 8 to match max. of Col 10

Col 13:  $S^{\text{LIF}}$ , normalised to max. = 1 [used for plotting]

Col 14:  $S^{\text{CELIF}}$ , normalised to max. = 1 [used for plotting]

## Data of figures in the Supplementary Material

### Figure S1: LIF and CRD transients

File: FigS1a TransOn\_ResBPEB-LIF.csv

Col 1: time after Nd:YAG Q-switch trigger [s]

Col 2: LIF PMT signal [V]

File: FigS1b TransOn\_ResBPEB-CRD.csv

Col 1: time after Nd:YAG Q-switch trigger [s].

Note that one significant figure is missing; the first data point is at  $-4.800\text{E-}7$  s and increases in increments of  $4\text{E-}10$  s, leading to the same time axis as in the file above.

Col 2: LIF PMT signal [V]

### Figure S2: Numerical vs fitted integral

File: FigS2\_135\_S1\_380\_traces.txt

Naming convention as for Fig. 5: BPEB molecular beam, nozzle temperature 135 °C (highest density), nozzle timing optimised to coincide with laser pulse, on resonance at 319.69 nm, LIF PMT voltage at 380 V.

Contains 101 CRD transients of 1000 time bins each.

Col 1: CRD PMT signal [V]

Col 2: [unused] (LIF PMT signal [V])

The time axis needs to be added manually:  $t = (i - 265) * 0.008 \mu\text{s}$ , in which  $i$  is the index of the if the time bin ( $i = 0 \dots 999$ ). Time bin  $i = 265$  corresponds to onset of the laser pulse entering the cavity and is equated to  $t = 0$ .

Each transient is separated by a blank line followed by the comment line "#CRDS[ tab] CeLIF"

### Figure S3: Ring-down time distributions

File: FigS2\_135\_S1\_380\_traces.txt

Same source as for Fig. S2 above.

File: FigS2\_075\_B1\_800\_traces.txt

Background measurement: BPEB molecular beam, nozzle temperature 75 °C (lowest density), nozzle mistimed (molecular beam pulse after laser pulse), on resonance at 319.69 nm, LIF PMT voltage at 800 V.

Same file format as for Fig. S2 above.

### Table S1

File: Fig03b\_N2-Rayleigh.csv

Same input data as Fig. 3(b) of the main paper.

### Figure S4: Ring-down time distributions

Files: FigS2\_135\_S1\_380\_traces.txt [same input data as for Figs S2 and S3].

FigS4\_135\_B1\_380\_traces.txt

FigS4\_135\_S0\_380\_traces.txt

FigS4\_135\_B0\_380\_traces.txt

Naming convention as for Fig. 5 of the main paper:

Nozzle temperature 135 °C (high BPEB density), LIF PMT at 380 V (low gain).

S1 is on resonance (319.69 nm) with nozzle timing optimised, the other three files are the various background measurements, see description of Fig. 5, above.