時間相関単一光子計数法を用いた原子、分子、クラスターの光励起実験

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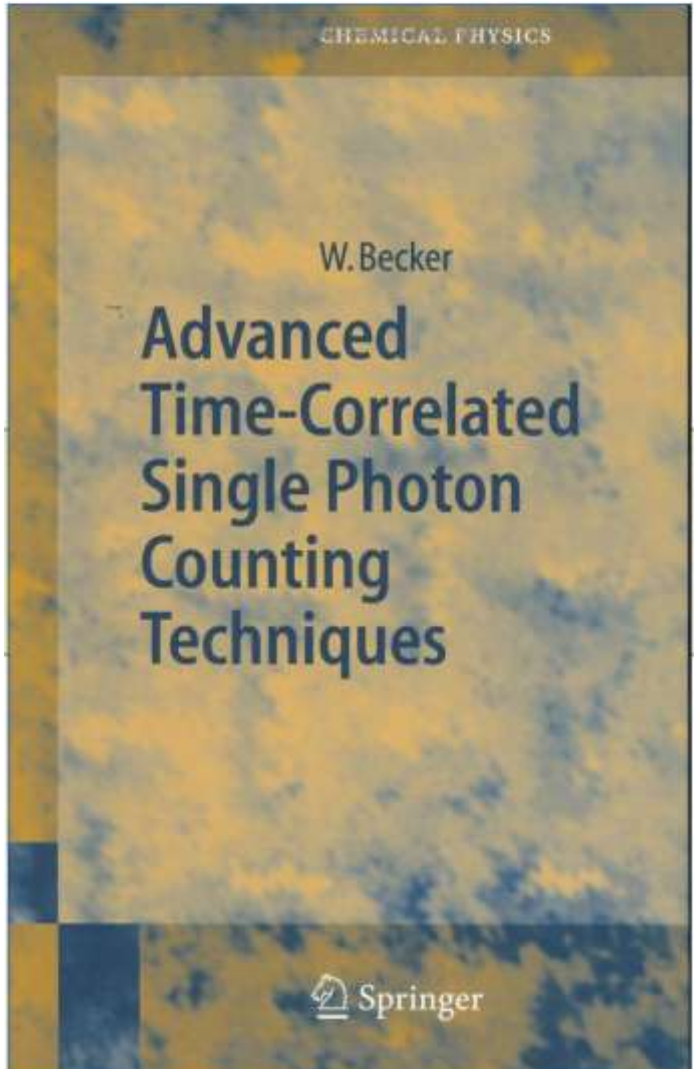
(SPring-8)

Well-established technique



Overview of TCSPC
Multidimensional TCSPC
Applications
•Fluorescence liftimes
•Time-resolved fluorescence
•Fluorescence lifetime spectrometers
•Fluorescence depolarisation
•Optical tomography
•Autofluoresence of biological tissue
•Laser scanning microscopy
•Fluorescence correlation
Detectors (PMT, MCP, avalanche)
Excitation sources (lasers)

Excitation sources

Table 7.1.

Light Source	Wavelength Range nm	Pulse Width ps	Rep. Rate (typ.) MHz	Power (CW) mW	Cost	Maintenance and Alignment Effort
Diode Laser	375, 405, 440, 475	50 to 300	0 to 80	0.2 to 2	low	none
Diode Laser	635, 650.... 1300	30 to 300	0 to 80	0.2 to 10	very low	none
Ti:Sa Laser	700 to 980	0.2 to 2	78 to 90	200 to 1300	high	low
Ti:Sa, Pulse Picker	700 to 980	0.1 to 1	0 to 9	<100	very high	high
Ti:Sa, SHG	350 to 490	0.1 to 1	78 to 90	100	high	medium
Ti:Sa, THG	250 to 320	0.1 to 1	78 to 90	20	high	medium
Ti:Sa, OPO	1050 to 1600	0.1 to 1	78 to 90	40 to 240	very high	high
+SHG	525 to 660	0.1 to 1	78 to 90	60 to 200	very high	high
Dye Laser	400 to 900	10	80 to 125	50	high	very high
Nd-YAG Laser	1064,	15	50 to 80	10,000	high	low
+ SHG, THG	532, 354, 266	15	50 to 80	100 to 4000	high	medium
Chip Laser	1064, 532, 354, 266	1500	< 0.01	20 to 1	low	none
Fibre Laser	800, 1600	0.2	80	20	medium	none
Synchrotron	X-Ray to IR	>1000	5	< 1	very high	very high

<0.1 for XFEL!

Lifetime of He 2*p*3*d*

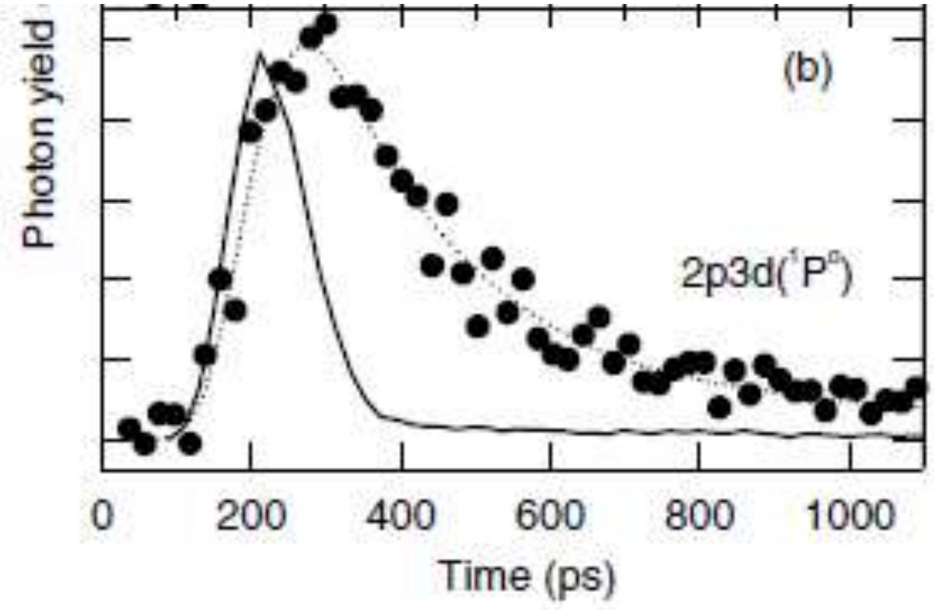


FIG. 1. (a) Photon yield from the He^+ $2p$ ion state and fit giving a lifetime of 102 ± 15 ps. (b) Photon yield from the $2p3d(^1P^0)$ doubly excited state and fit giving a lifetime of 190 ± 30 ps. The solid lines are instrumental response functions of the system.

He** lifetimes at PF

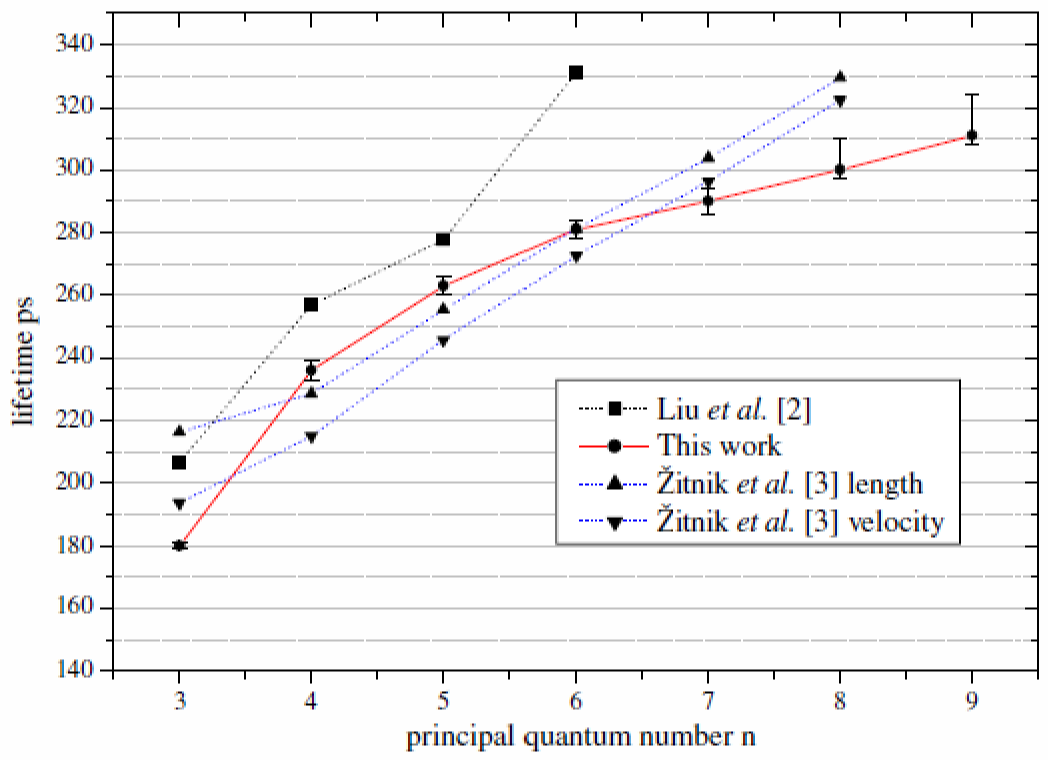
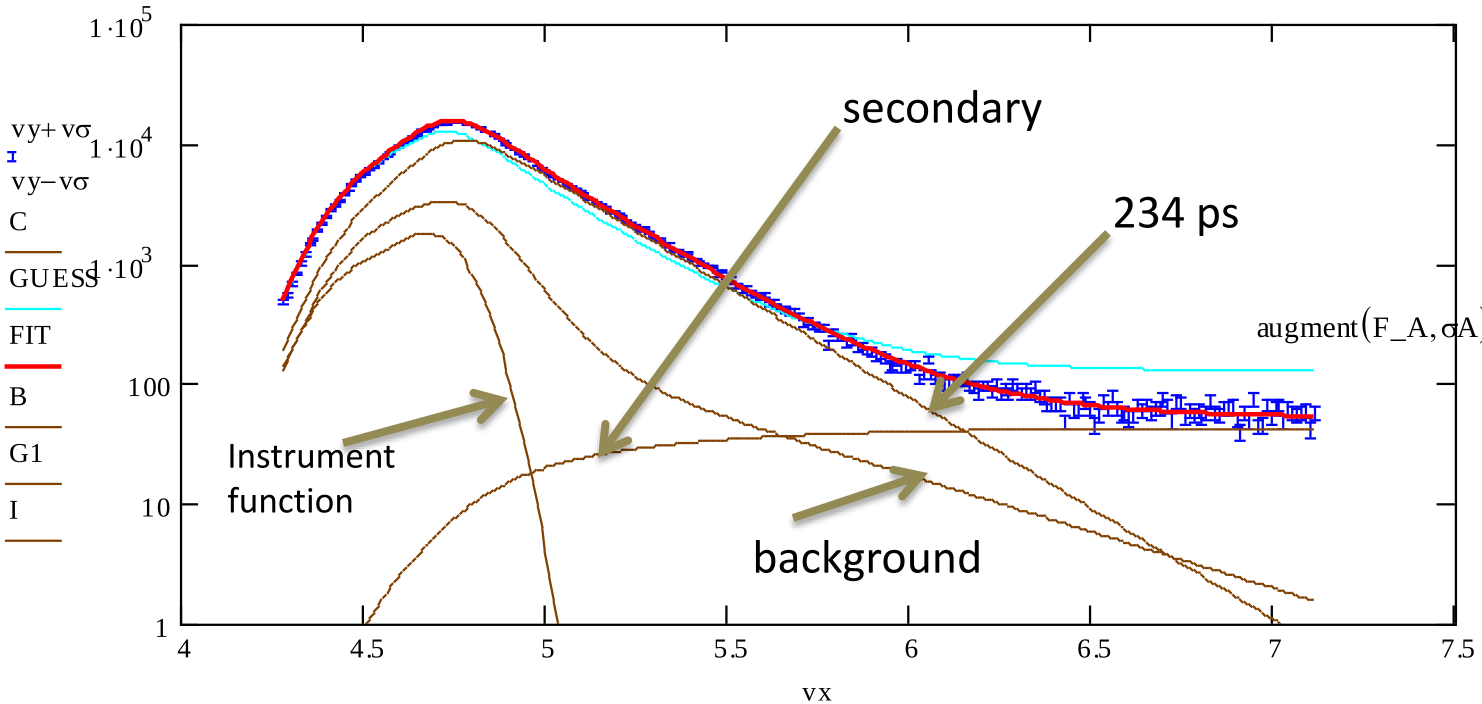


Figure 2. Comparison of the experimental $(2,-1)_n$ lifetimes with the theoretical results of Liu *et al* [2] and Žitnik *et al* [3]. The points are joined with straight lines for clarity only.

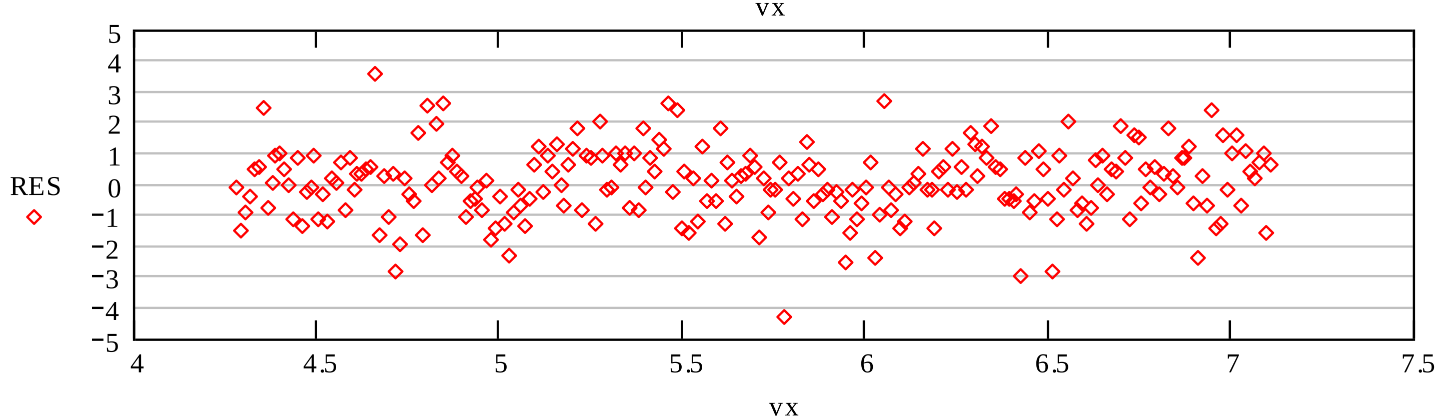
Fitting: 2*p*4*d*



$$\text{augment}(F\_A, \sigma A) = \begin{pmatrix} 18.3336 & 0.4966 \\ 3121.4358 & 7.6025 \\ -0.0188 & 0.0004 \\ 0.0857 & 0.0007 \\ 4.68 & 0.0006 \\ 0.1254 & 0.0006 \\ 4.5057 & 0.0006 \\ 234.7685 & 0.6422 \\ 0.7037 & 0.0034 \\ 2.2408 & 0.0198 \\ 1380.7171 & 22.5555 \\ 7.6794 & 1.1147 \end{pmatrix}$$

$\text{CHISQV}(F\_A) = 1.3413$

$\text{PROB}(F\_A) = 24.6802$



Lifetime-resolved spectroscopy

Photodissociation dynamics of a hydrogen molecule via superexcited states as studied through the angular-momentum population of an excited hydrogen atom

Norihisa Terazawa,[a] Noriyuki Kouchi, Masatos
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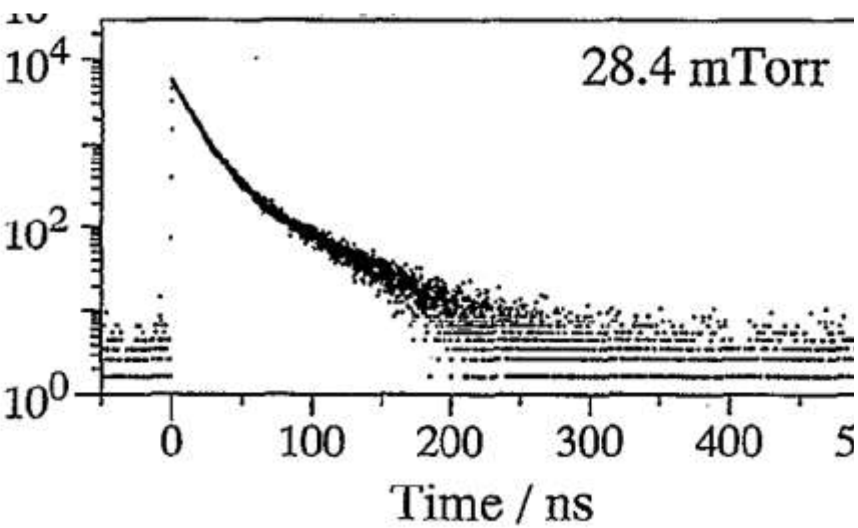
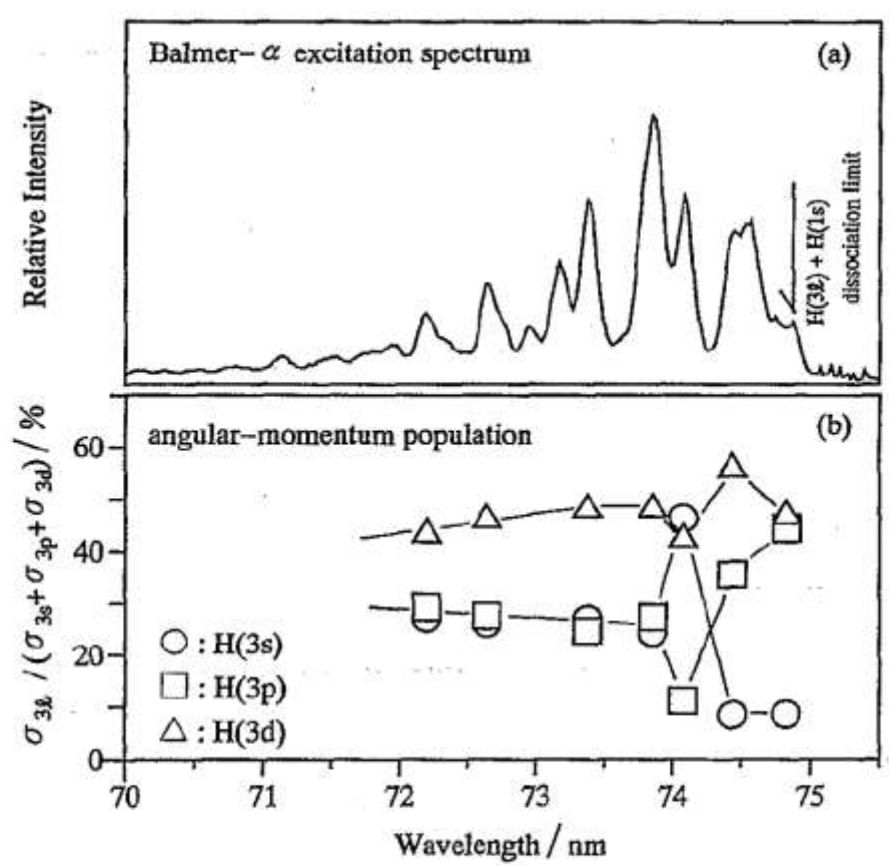
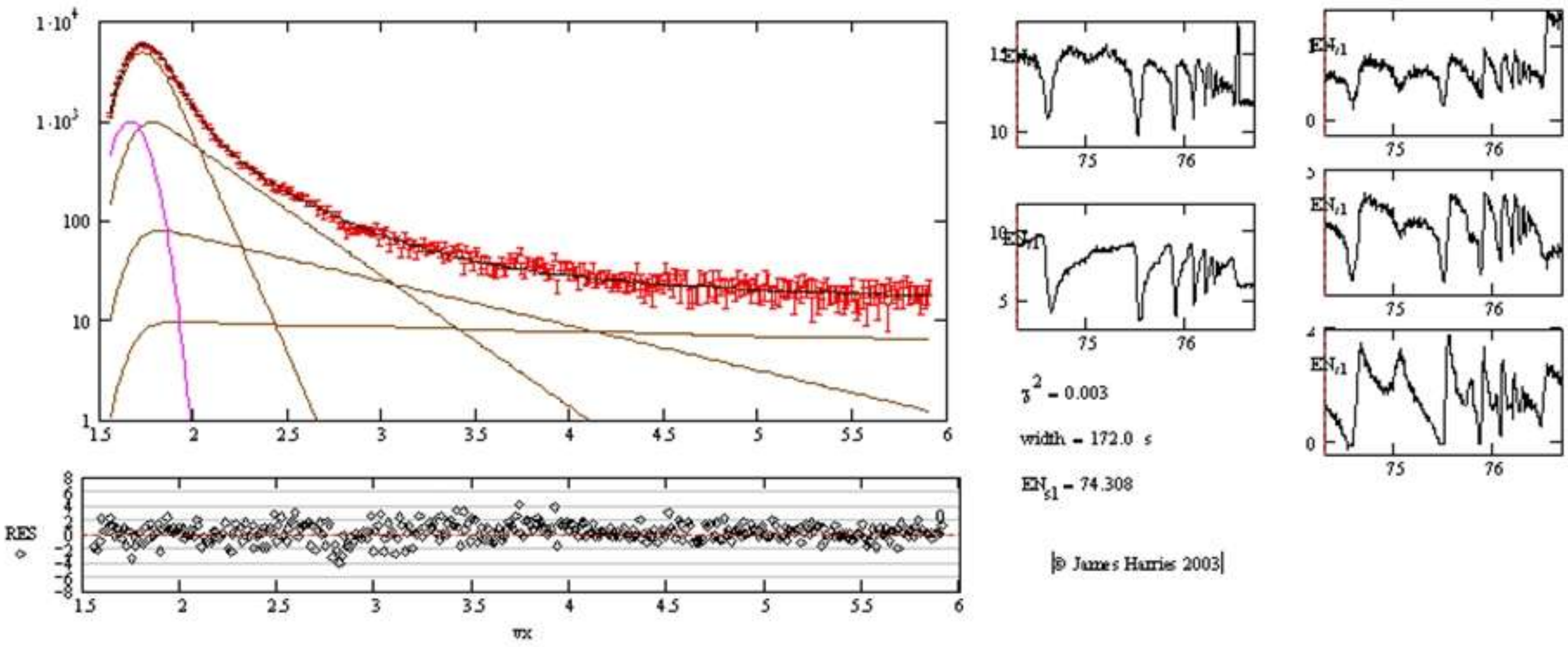
Kenji Ito
Photon Factory, National Laboratory for High Energy

7036 J. Chem. Phys. 100 (10), 15 May 1994 0021-96





He^{+} LRFS continuous scan @PF



Argon Clusters

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Investigation of the 2p ionization threshold region of Ar clusters by observation of the fluorescence lifetime

T. Gejo [a,*], M. Iseda [a], T. Tamura [a], K. Honma [a], J.R. Harries [b], Y. Tamenori [b]

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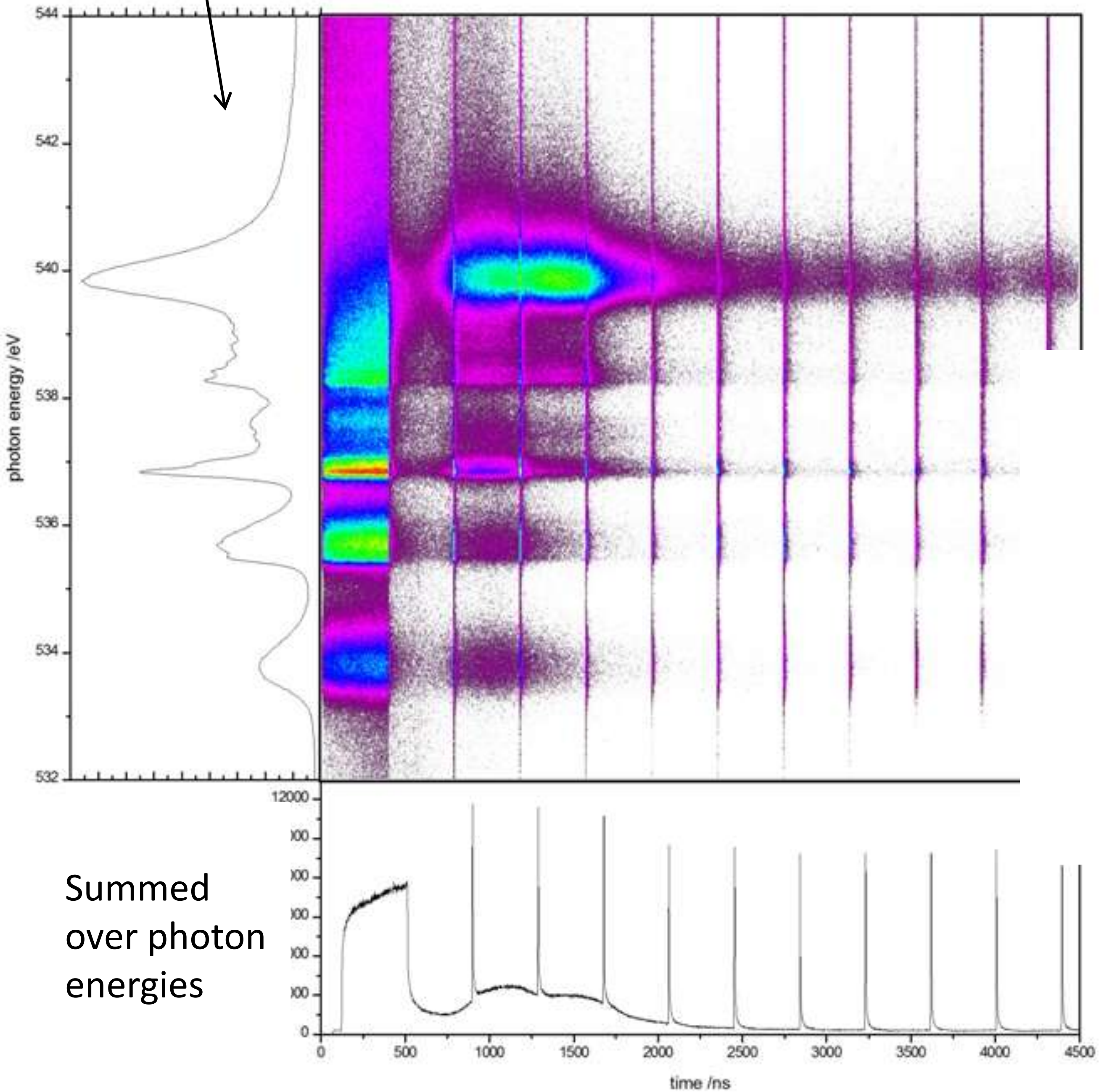
Available online 26 November 2006

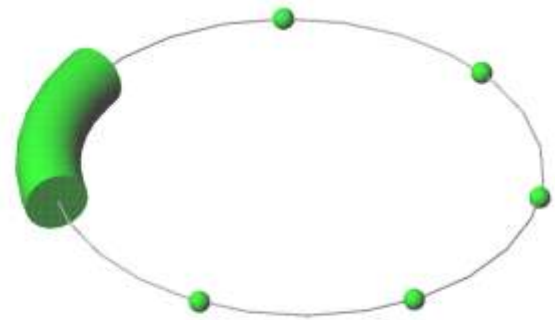
Counts, summed over all arrivals times == TFY+TMY



Summed over photon energies

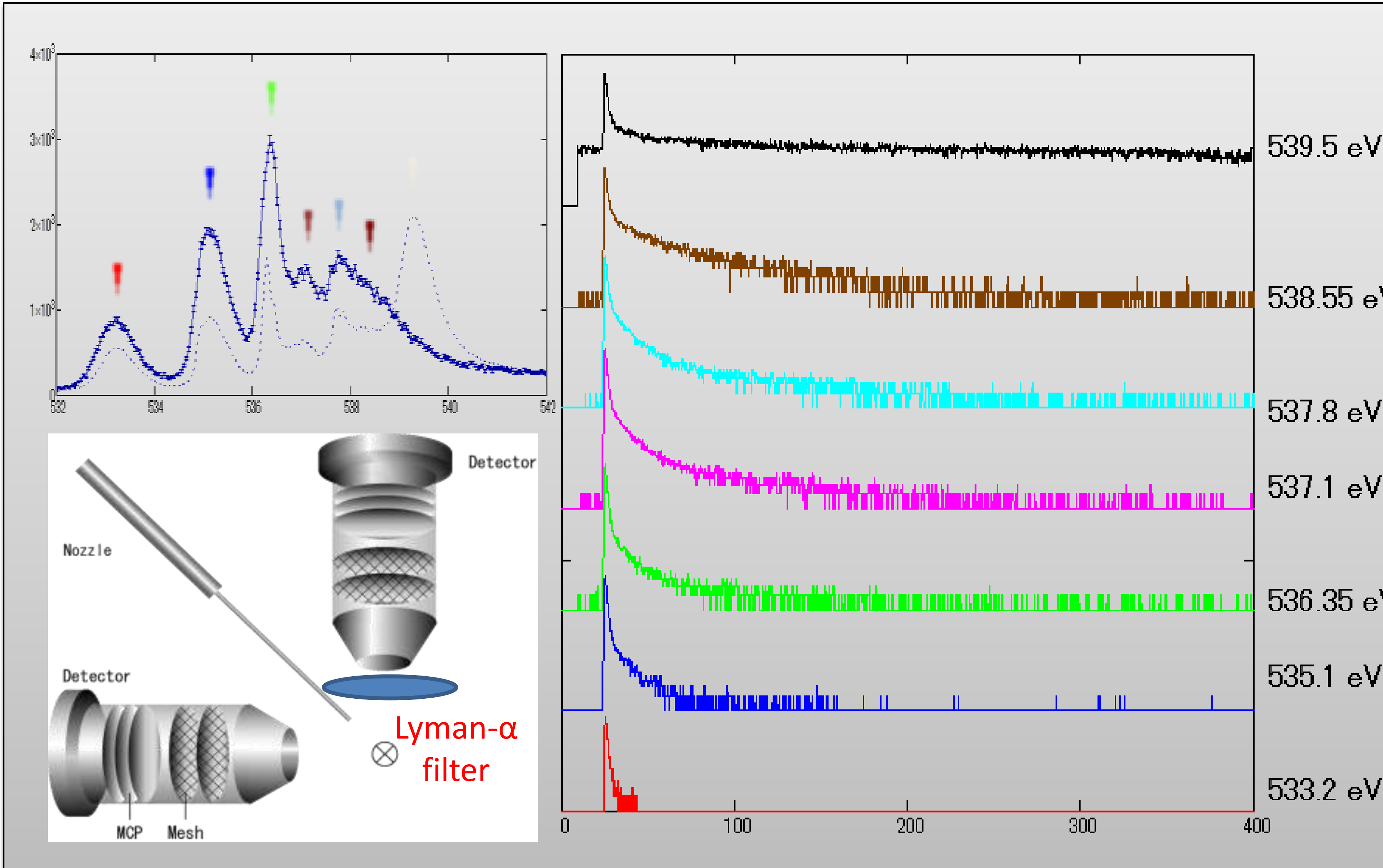
Neutrals arrival times vs. photon energy.

D-mode



D-mode:
"1/14+12" filling
1.6mA SBx12 (342ns)
80 mA of MB in 342ns

H(2p→1s)　蛍光寿命



Detector
Nozzle
Detector
MCP
Mesh
Lyman-α
filter
539.5 eV
538.55 eV
537.8 eV
537.1 eV
536.35 eV
535.1 eV
533.2 eV
0
100
200
300
400

LRFS future: imaging?

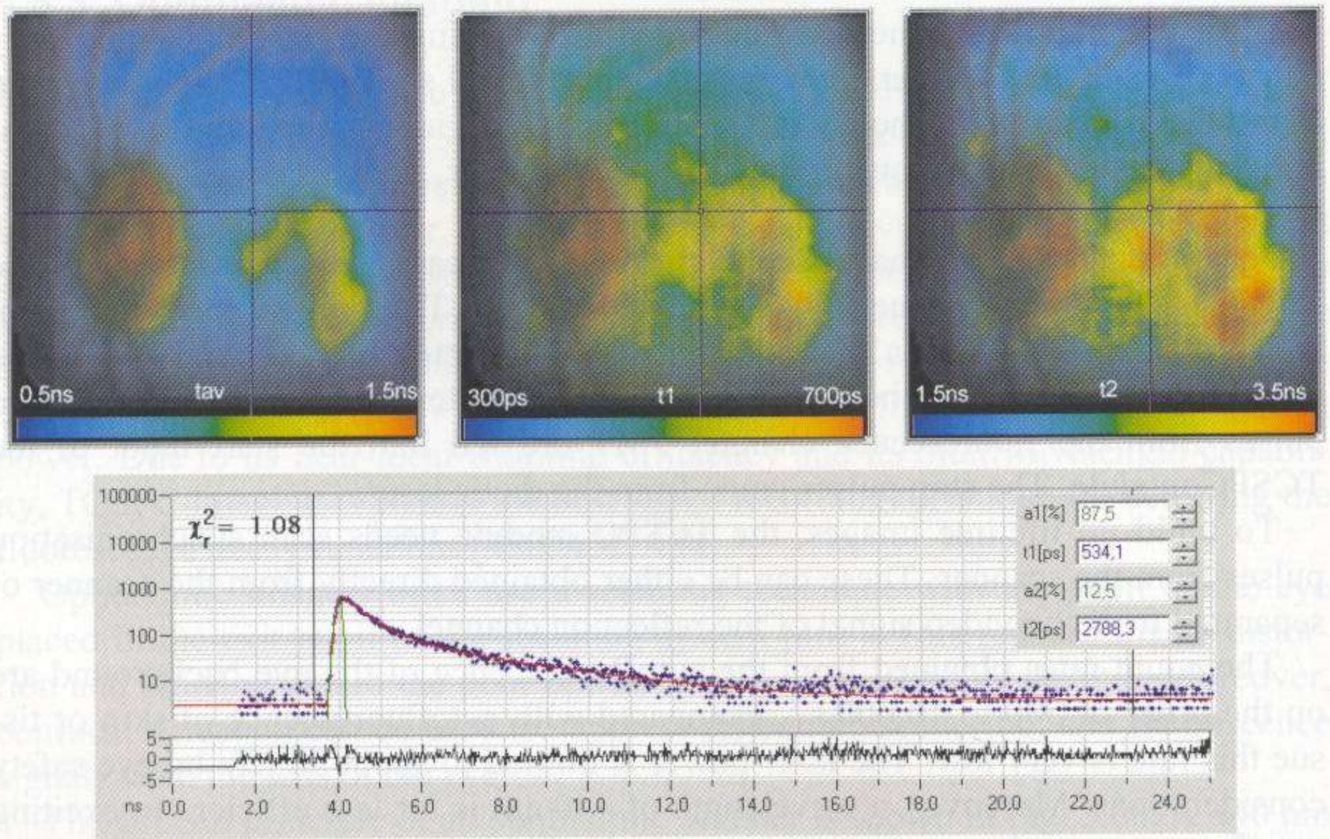


Fig. 5.69 TCSPC lifetime images of the human ocular fundus, obtained by a double-exponential fit. The lifetime is colour-coded as indicated in the images. Upper *left*: Average lifetime. Upper *middle*: Fast lifetime component. Upper *right*: Slow lifetime component. Lower part: Decay curve in the selected spot and fit results. Courtesy of Dietrich Schweitzer, Friedrich Schiller University Jena, Germany

Lifetime depends on concentration of O_2