<u>Quantum entangled</u> two-photon absorption for selective, localized, and low intensity pumping of excited state populations in plasma

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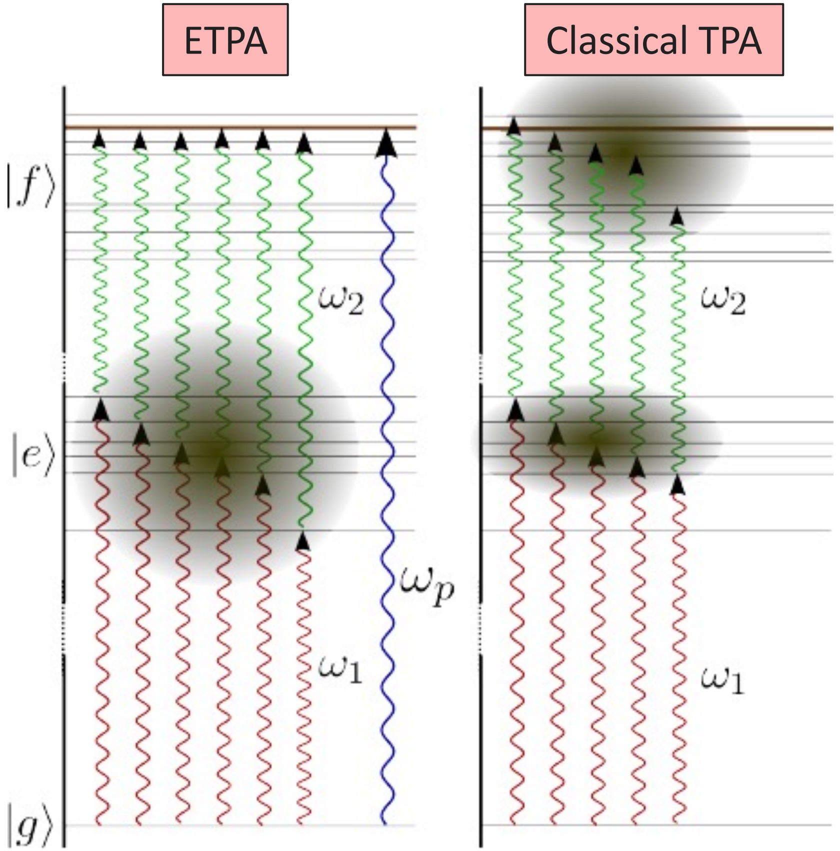
Entangled two-photon absorption for plasma spectroscopy

- Entangled two-photon absorption (ETPA) may enable …
  - Efficient pumping of an excited state population with a modest-power CW laser
  - High-bandwidth measurements of the fluorescing population
- Here, we explore the feasibility of ETPA in plasma and a plan for proof-of-principle measurements
  - Localized density measurements at MHz timescales
    - CX products, partially-ionized impurities (intrinsic or injected), neutrals (edge or beam)

Classical vs. entangled two-photon absorption (TPA)

Entangled TPA (ETPA)

- Simultaneous generation and arrival of entangled photon pair (EPP)
  - EPP generation and ETPA is a single random process
  - ETPA cross-section scales linearly with laser intensity
  - 10^4-10^7 quantum enhancement allows for low power CW laser
- Sum frequency (total energy) retains linewidth (presumably narrow) of the laser source
  - $\omega_p = \omega_1 + \omega_2$
  - Avoids excitation of non-target states



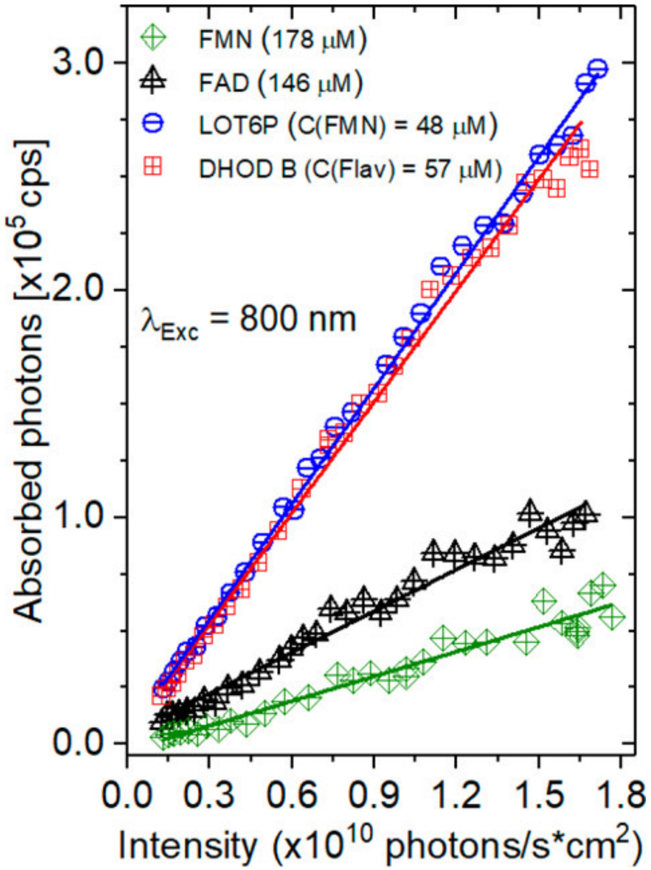
Schlawin et al, Phys. Rev. A 86, 023851 (2012)

Classical TPA

- Each photon absorption is an uncorrelated random process
  - TPA cross-section scales quadratically with laser intensity
  - Requires high power laser for observable TPA ➞ short-pulse laser with broad linewidth
- Broad linewidth lacks specificity for the target excited state
  - Spurious excitation of non-target states
- Also known as TALIF (two-photon absorption laser induced fluorescence)

ETPA and classical TPA cross sections

ETPA



ETPA rate scales linearly with $\boldsymbol{\phi}$

Classical TPA scales quadratically with $\boldsymbol{\phi}$

$$R_E = \frac{1}{2}(\sigma_E \phi + 3\sigma_C \phi^2)$$

R_E = total TPA rate [1/t]

$\boldsymbol{\phi}$ = photon number flux [1/d^{2}t]

σ_C = classical TPA cross section [d^{4}t]

σ_E = entangled TPA cross section [d^2]

$$\sigma_E \approx \frac{\sigma_C}{T_e A_e}$$

T_e = entanglement time [t]

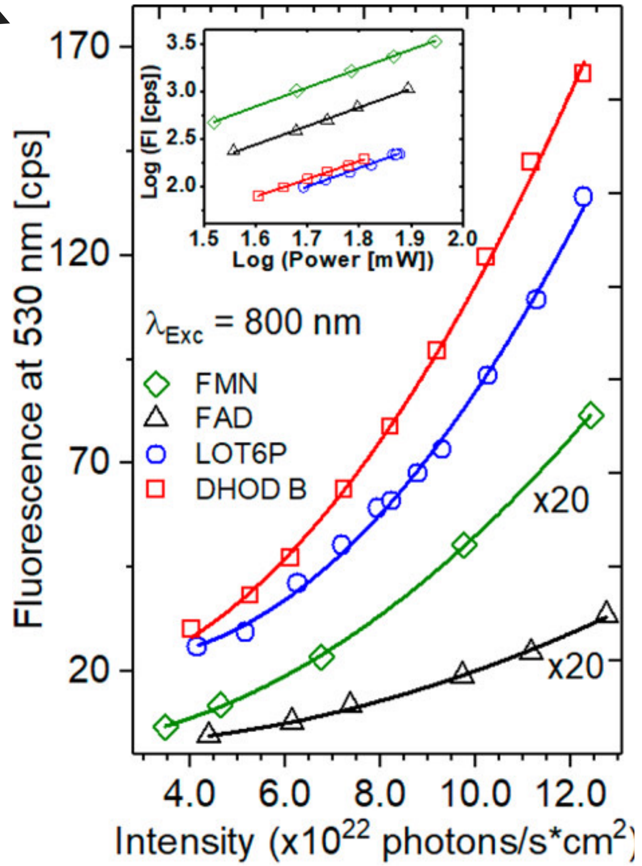
$B_e = 1/T_e$ = entanglement bandwidth [1/t]

A_e = entanglement area [d^2]

For short T_e (large B_e) and small $\boldsymbol{\phi}$:

$$\sigma_E \boldsymbol{\phi} >> \sigma_C \boldsymbol{\phi}^2$$

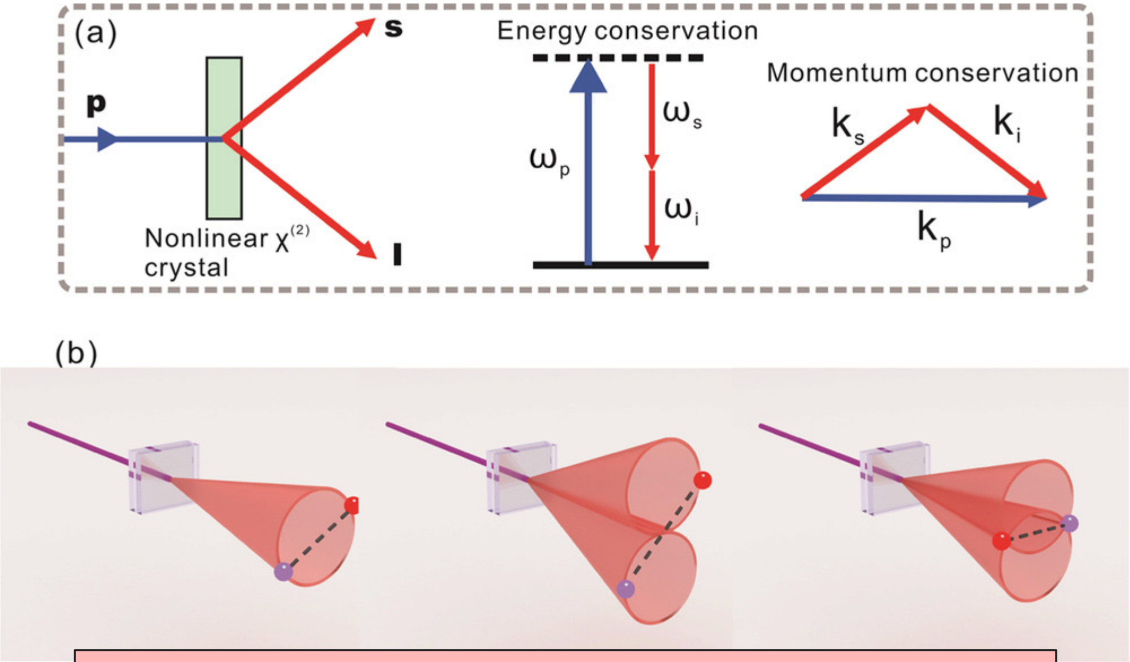
Classical TPA



Parzuchowski et al, Phys Rev Appl 15, 044012 (2021)
Villabona-Monsalve et al, J. Am. Chem. Soc. 140, 14562 (2018)

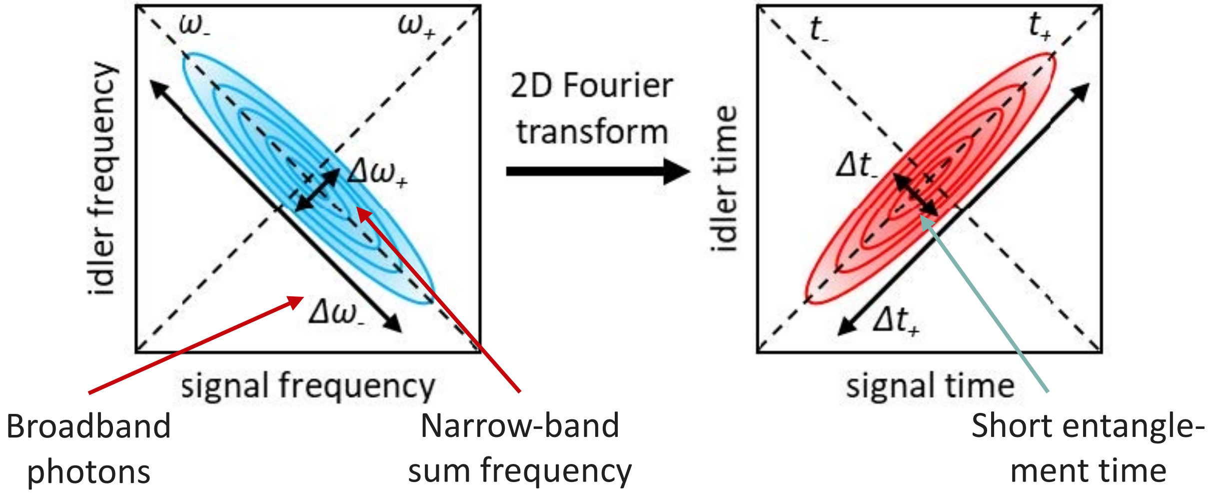
Entangled photon pair generation and time-frequency entanglement

Spontaneous parametric down-conversion (SPDC)



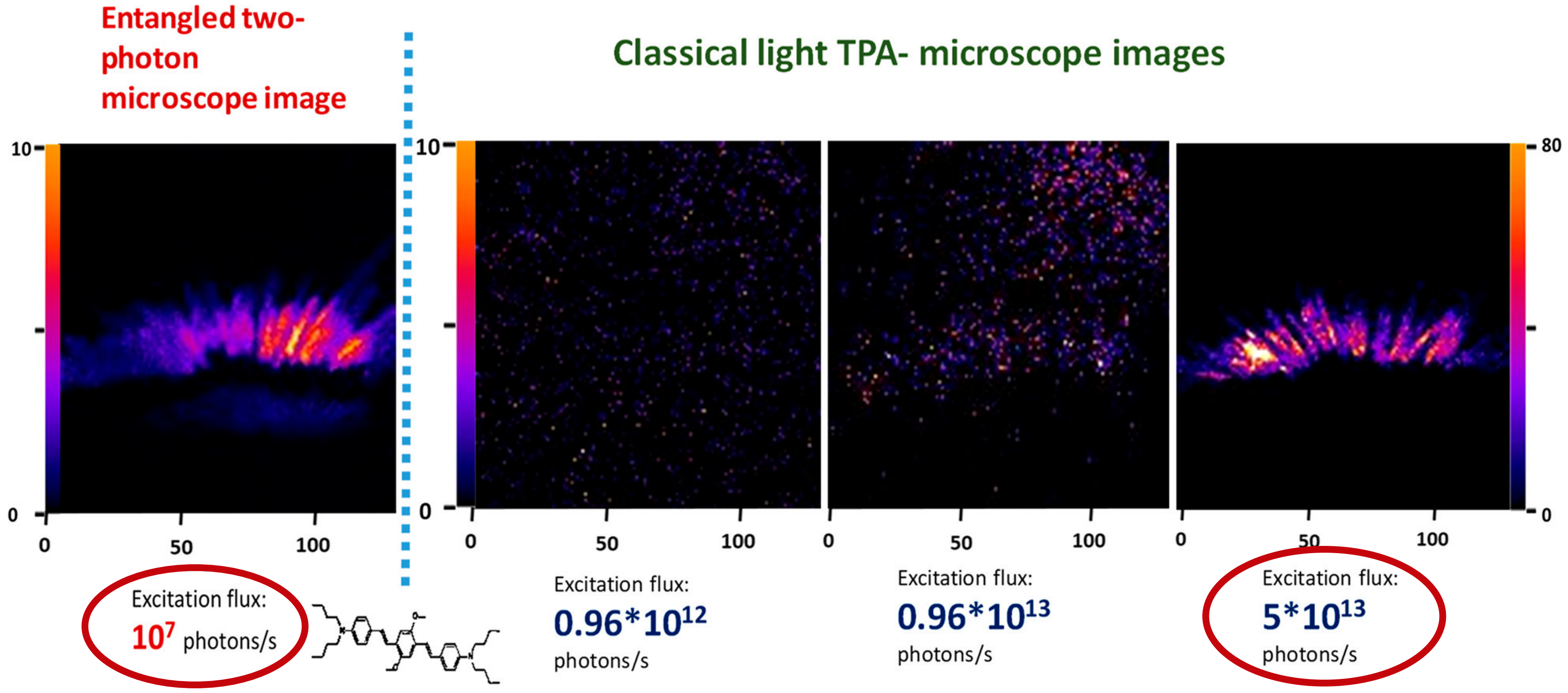
The SPDC "type" characterizes the EPP polarization, direction, and frequency. These characteristics are set by the orientation of the probe beam and nonlinear crystal.

- Single (decohered) photons have time and energy (frequency) Fourier pairs and are subject to a time-energy uncertainty limit.
- Entangled photons pairs (EPP) have Fourier pairs for sum and difference time and energy such that the delay time and sum frequency are not bound by an uncertainty limit. Fourier pairs of the EPP system:
  - Entanglement time and photon bandwidth: Δt- ⇆ Δω-
  - EPP creation time and sum bandwidth: Δt+ ⇆ Δω+



Zhang et al, Adv. Quant. Tech. 4, 2000132 (2021)
R. Burdick, Ph.D. Thesis, U. Mich., 2021

Quantum enhancement with ETPA

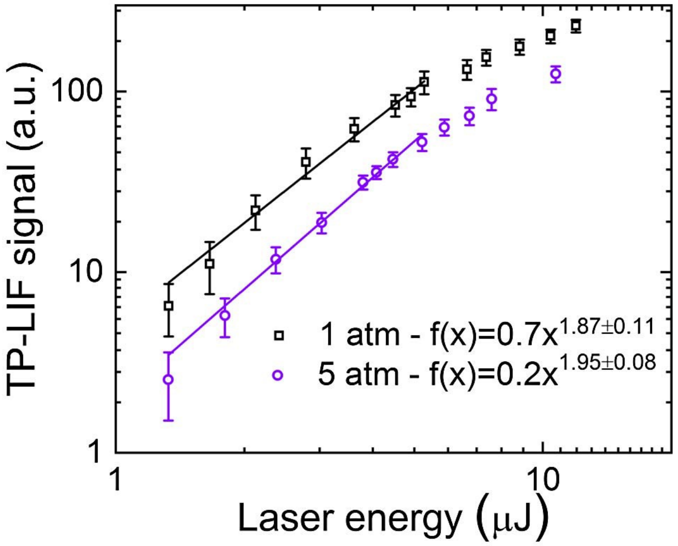


- ETPA fluorescence is comparable to classical TPA with a factor of $\mathbf{10^6}$ **fewer photons**
- Note: The order-of-magnitude for the quantum enhancement is an active area of debate, and estimates are in the range 10^4-10^7.

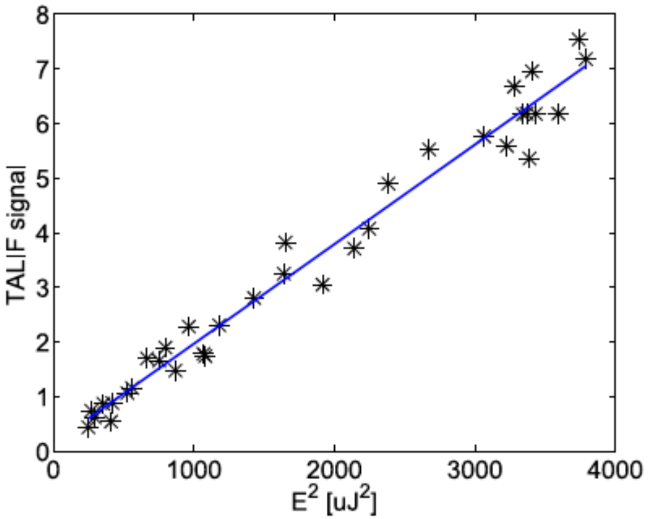
Eshun et al, Acc. Chem. Res. 55, 991 (2022)

Classical TPA with high-intensity, pulsed laser shows the quadratic scaling with laser intensity

Reference lines show quadratic scaling for classical TPA with laser intensity

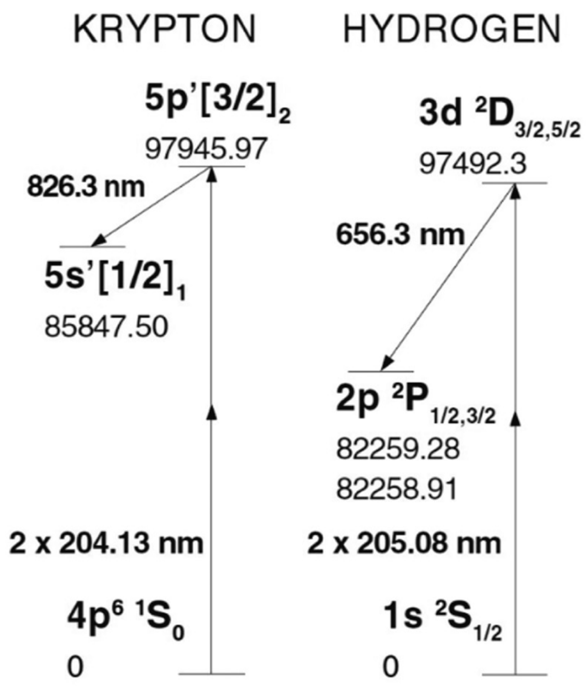


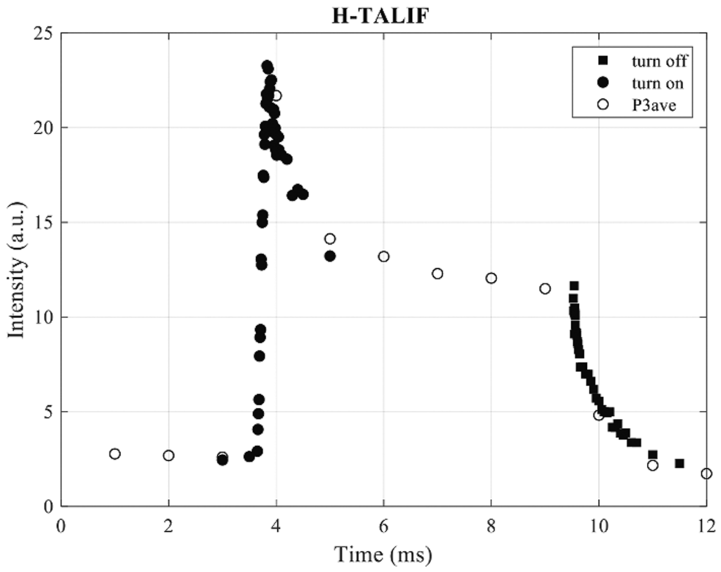
- Oxygen (O I) TPA in flame
- 2×225.6 nm for 2p (gnd state) ➞ 3p
- Fluorescence at 845 nm
- Ti:Sa with 100 fs pulses @ 1 kHz; pulses with 40 μJ and irradiance ~10^{11} W/cm^2
- Rahman et al, Applied Optics (2019)



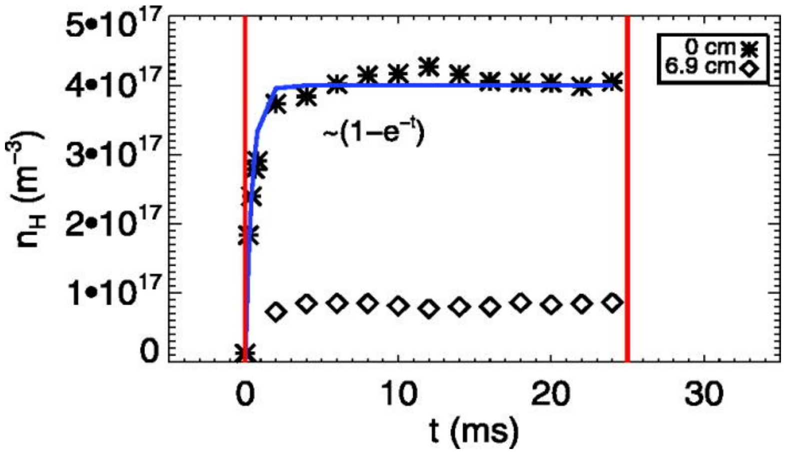
- H TPA in dielectric barrier discharge
- 2×205 nm for 1s (gnd state) ➞ 3p
- Fluorescence at 656 nm
- YAG pumped dye laser; 8 ns pulses at ~100 μJ and irradiance ~10^{12} W/m^2
- Dvořák et al, Plasma Sources Sci. Technol. (2017)

Classical TPA in plasma: H/D density measurement with ground state excitation





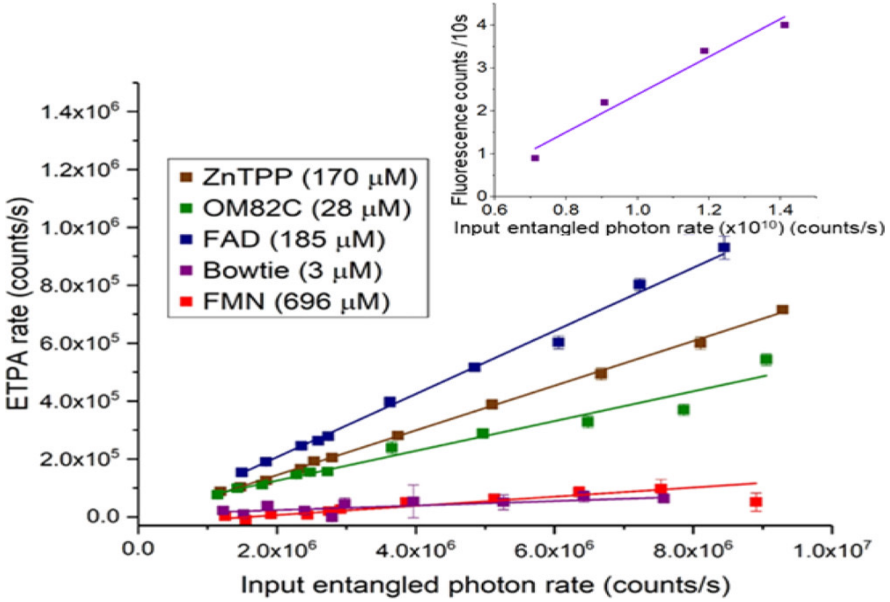
- Ti:Sa @ 1 kHz; 100 fs and 0.1 mJ per pulse at 205 nm
- Dogariu et al, RSI 93, 093519 (2022)



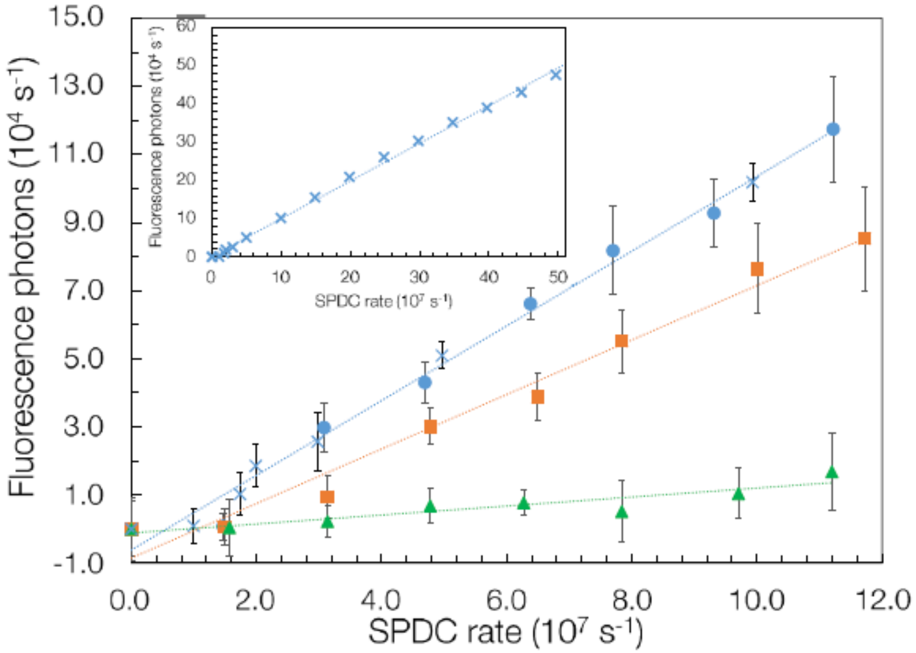
- Nd:YAG @ 20 Hz; 8 ns and 8 mJ per pulse at 205 nm
- Galante et al, Phys. Plasmas 21, 055704 (2014)

ETPA with CW laser sources

Reference lines show linear scaling for ETPA with laser intensity



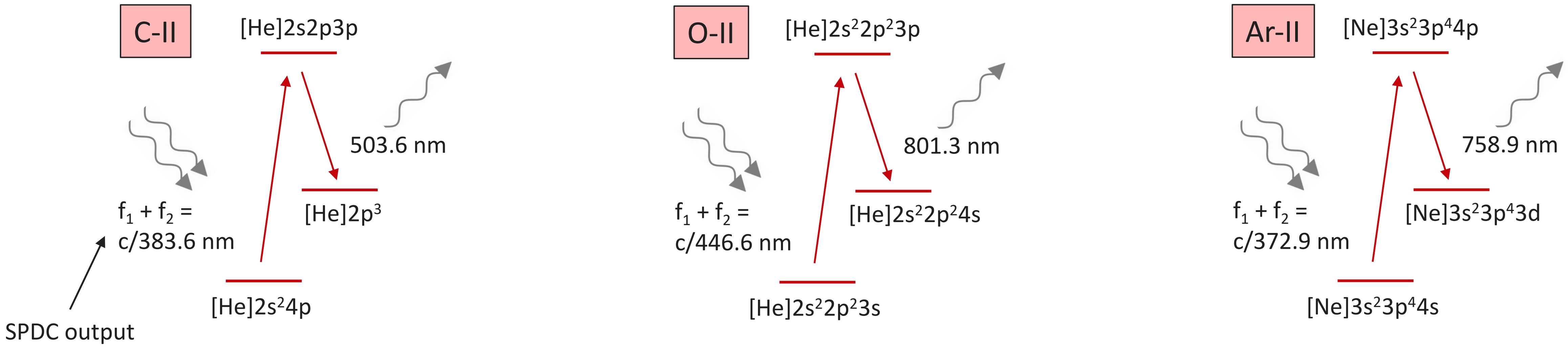
- CW diode laser, 70 mW
- Villabona-Monsalve et al, J Phys Chem C, 124, 24526 (2020)



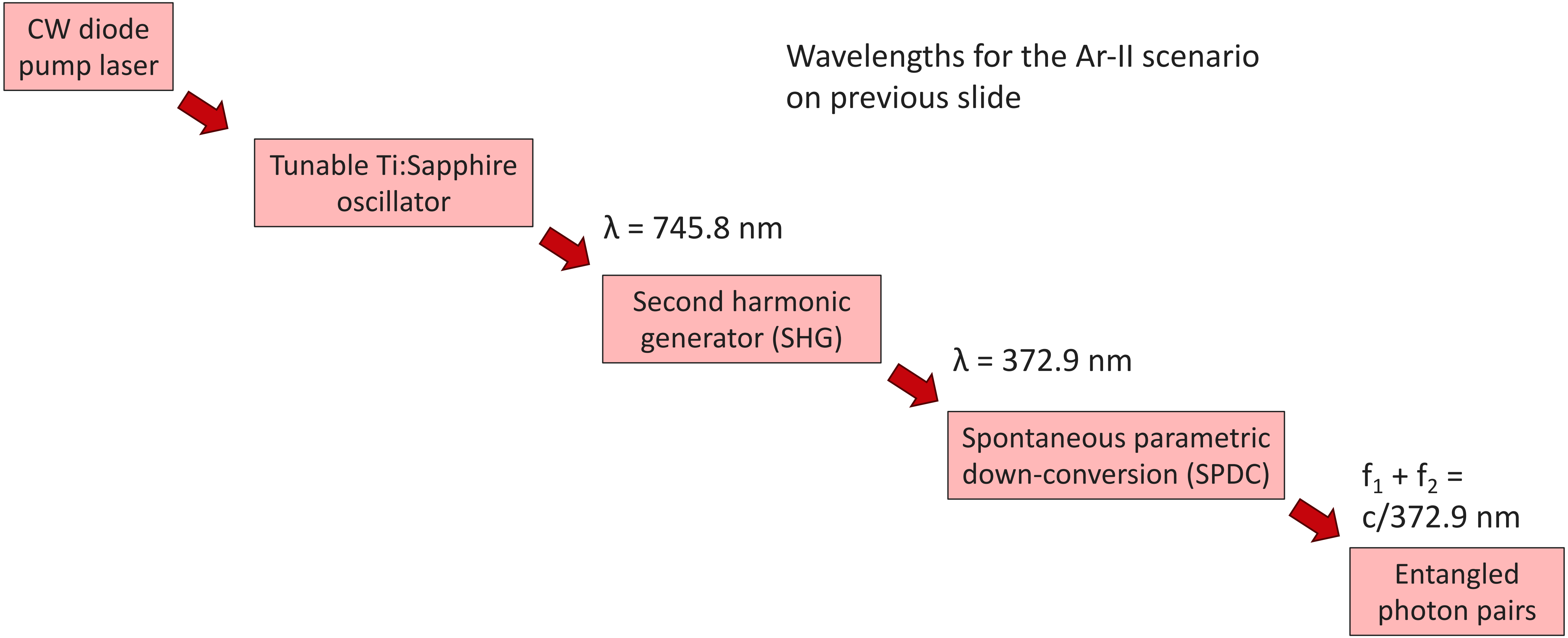
- CW diode laser, 5 W
- Tabakaev et al, Phys Rev A 103, 033701 (2021)

Pump and fluorescence transitions for a proof-of-principle demonstration in a ~1 eV helicon plasma

- Target pump transitions near 200-400 nm (3-6 eV)
  - ≳ 3 eV (≲ 400 nm) to minimize electron impact excitation
  - ≳ 200 nm (≲ 6 eV) to avoid VUV complexities
  - If feasible, ≳ 350 nm for compatibility with freq. doubled Ti:Sapphire laser, but 3f or 4f is feasible for shorter wavelength
- Target visible fluorescence (400-800 nm) for detection with fast response, low noise photodetectors



CW laser source and SPDC



Summary and outlook

- Entangled two-photon absorption (ETPA) may enable …
  - Efficient pumping of an excited state population with a modest-power CW laser
  - High-bandwidth measurements of the fluorescing population

- We will attempt to demonstrate ETPA in a ~1 eV helicon plasma
  - CW pumping of an Ar-II transition and detection of the fluorescence