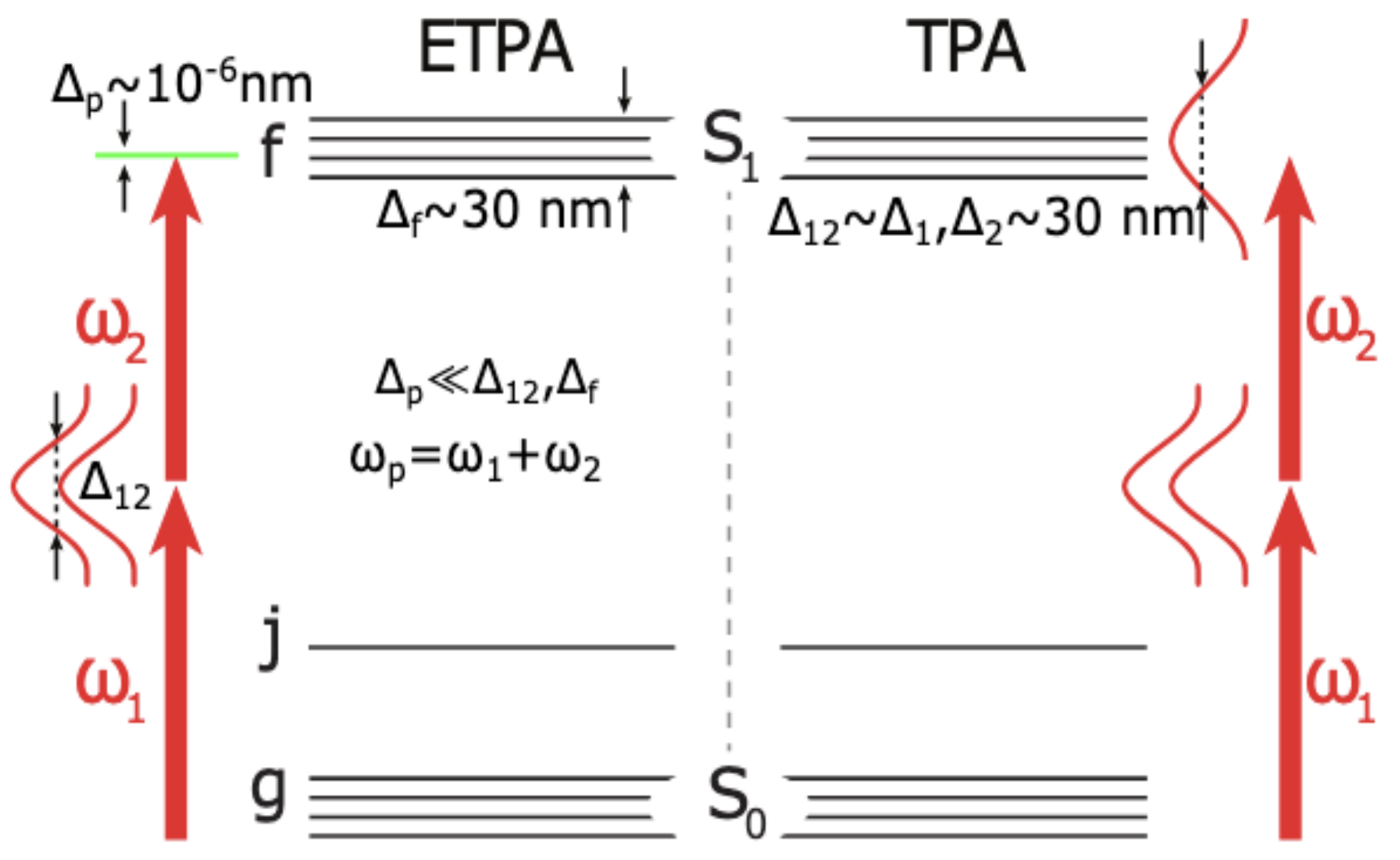
E²TPA: Exploiting Entangled Two-Photon Absorption

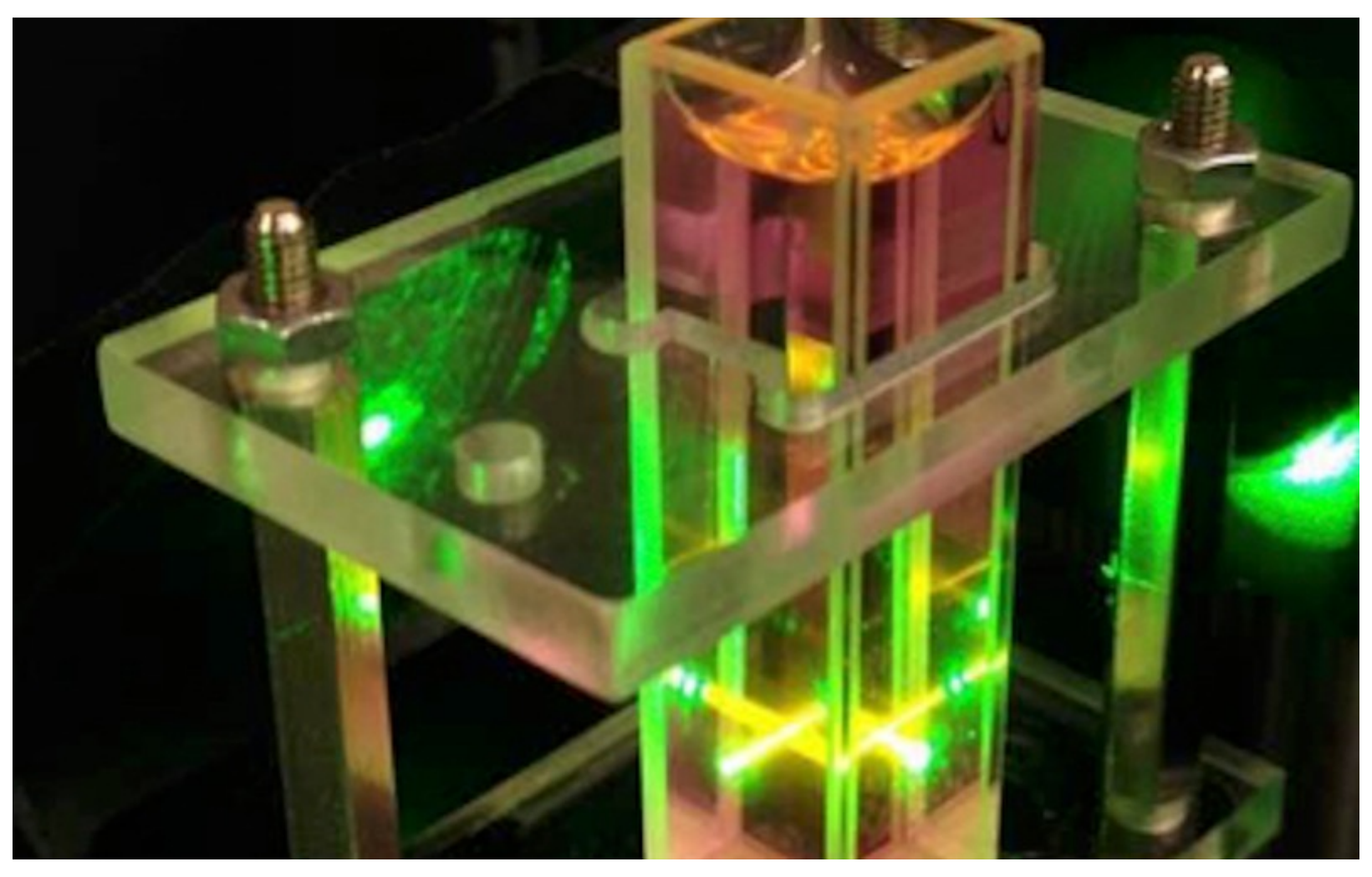
What is Entangled Two-Photon Absorption?

Entangled two photon absorption (ETPA) is analogous to classical two photon absorption (TPA), except in the case of ETPA, the two photons act as a single quantum object – always correlated and arriving at the molecule at the same time – behaving like the much more efficient single photon absorption. This effect represents one of the most significant quantum advantages with real application potential in the near-term.



E²TPA's Goals

- ❖ Develop cohesive theoretical & experimental guidelines for application design.
- ❖ Fabrication & development of quantum light sources optimised for ETPA systems.
- ❖ Design & exploitation of plasmonic nanoarrays for enhanced ETPA performance.
- ❖ Demonstrate ETPA providing a baseline for quantum-enhanced system design.

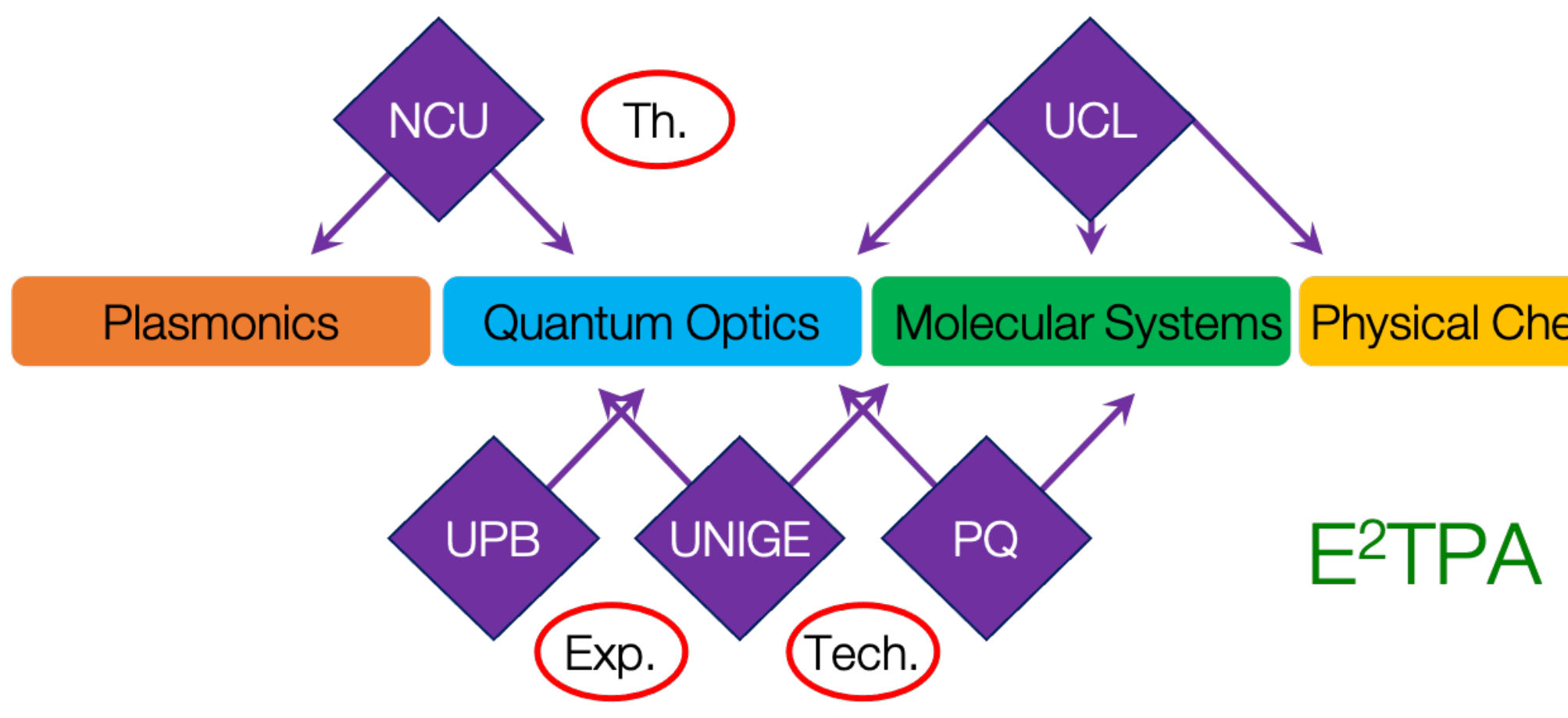


Before E²TPA	After E²TPA
Contradictory experimental results and theoretical predictability of theory limited.	Predictive theoretical model bridging existing conceptual approaches confirmed by experiments.
Light sources not optimised.	Dedicated light sources: brightness; bandwidth, and tuneability ETPA-optimised.
Proof-of-principle experiments for ETPA.	ETPA cross-sections and fluorescence characterised for a comprehensive set of experimental scenarios.
Preliminary theoretical predictions of plasmonic ETPA enhancement.	Plasmonic influence fully characterised for local & spatially integrated absorption fluorescence rates.

Motivation

Nonlinear spectroscopy and microscopy techniques are ubiquitous in a wide range of applications across physics and biology. However, these usually rely on high-powered pulsed laser systems. A promising alternative is to exploit entangled two-photon absorption (ETPA), which can lead to tens of orders of magnitude lower incident fluxes than in conventional two-photon absorption schemes.

Partners, Competencies & Expertise



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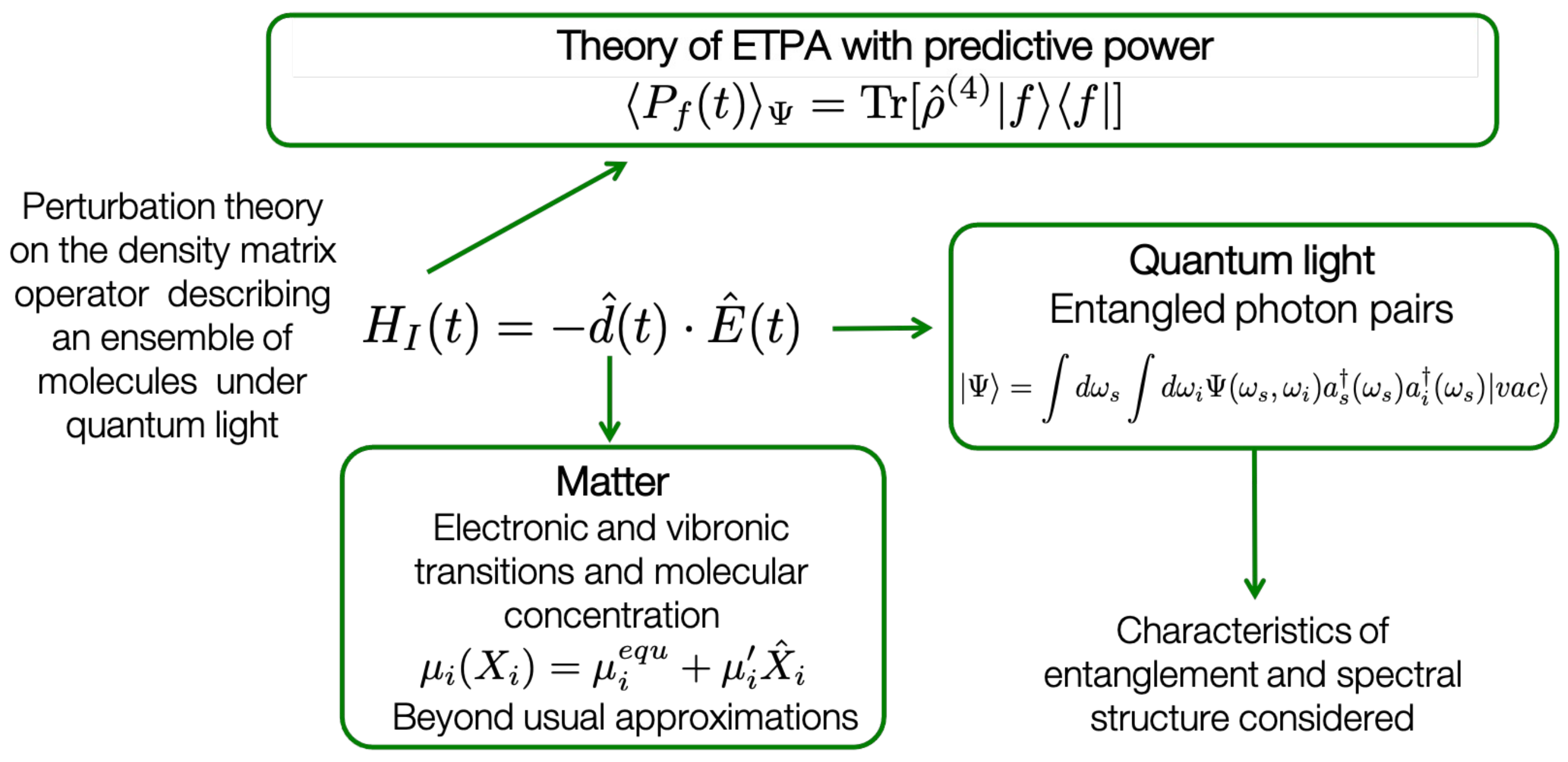
E²TPA



Theory of ETPA with predictive power
$\langle P_f(t)\rangle_\Psi = \mathrm{Tr}[\hat{\rho}^{(4)}|f\rangle\langle f|]$

Perturbation theory on the density matrix operator describing an ensemble of molecules under quantum light

$H_I(t) = -\hat{d}(t)\cdot\hat{E}(t)$

Quantum light
Entangled photon pairs
$|\Psi\rangle = \int d\omega_s \int d\omega_i \Psi(\omega_s,\omega_i) a_s^\dagger(\omega_s) a_i^\dagger(\omega_s)|vac\rangle$

Matter
Electronic and vibronic transitions and molecular concentration
$\mu_i(X_i) = \mu_i^{equ} + \mu_i'\hat{X}_i$
Beyond usual approximations

Characteristics of entanglement and spectral structure considered

Theory

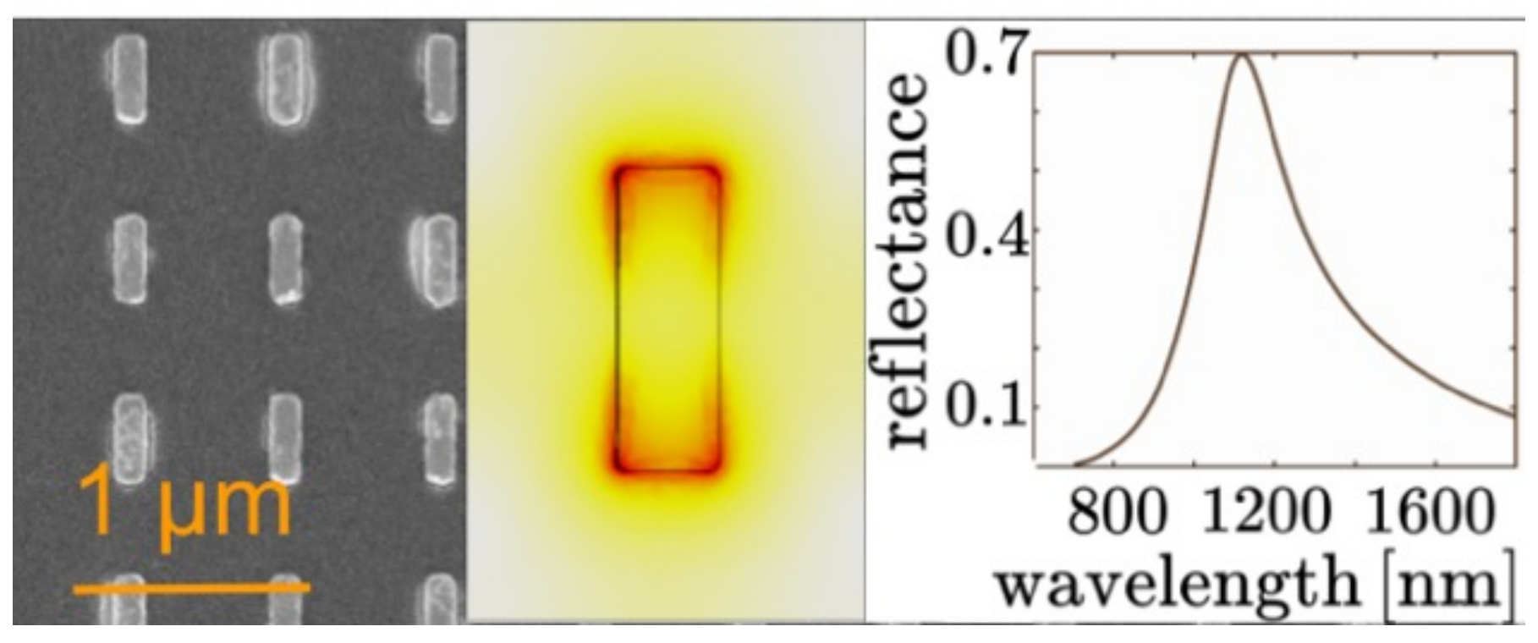
We take an holistic approach to better understand the complete light-matter system to determine and exploit the quantum advantage afforded by ETPA. In particular, a relationship between classical and entangle two photon absorption cross-sections has been identified:

$$\sigma_{ETPA} = f\,\frac{\sigma_c}{AT}$$

where σ_i represent the entangled and classical cross-sections respectively, and A and T are the entanglement area and time. The factor f has been introduced as an ad-hoc pre-factor to compare the quantum enhancement.

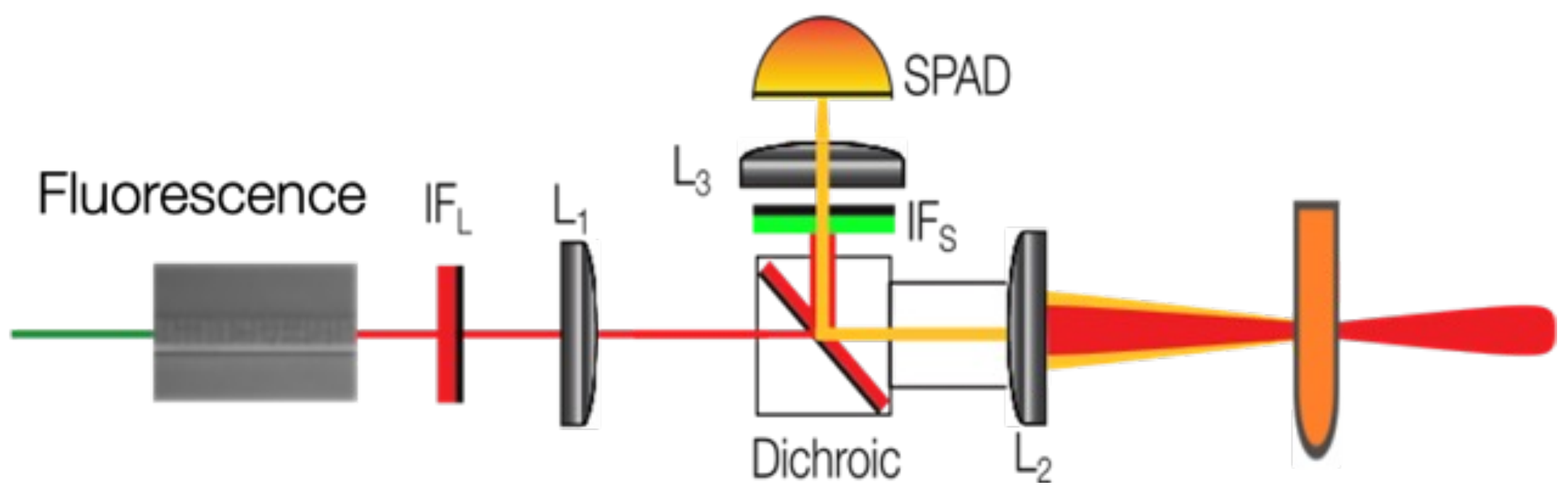
Plasmonic Nanoarrays

We will clarify the trade-off between the local field enhancement by the nanostructures and the absorption losses they sustain. We will engineer nanoarrays to maximise the ETPA performance for selected target molecules.



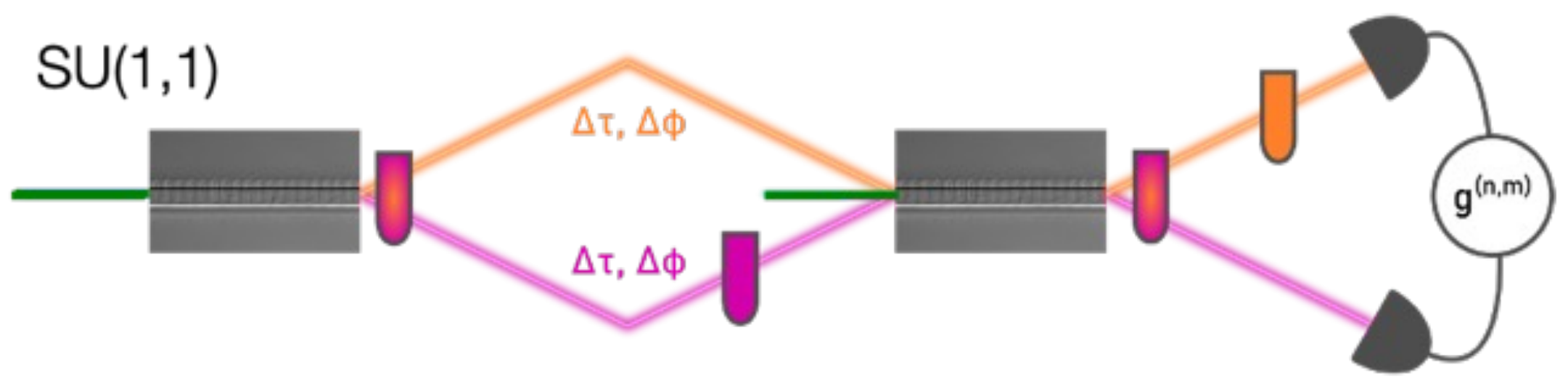
Fluorescence Schemes

Fluorescence detection schemes are closest to existing microscopy & spectroscopy approaches. We aim to improve the SNR and test theoretical proposals to better understand temporal and spatial characteristics and the light-matter interaction itself.



Transmission Schemes

This method is based on broadband correlation functions g(n,m). These are robust against linear loss but highly sensitive to correlated loss. We can also introduce a time delay between the two photons, reducing uncertainties from switching samples.

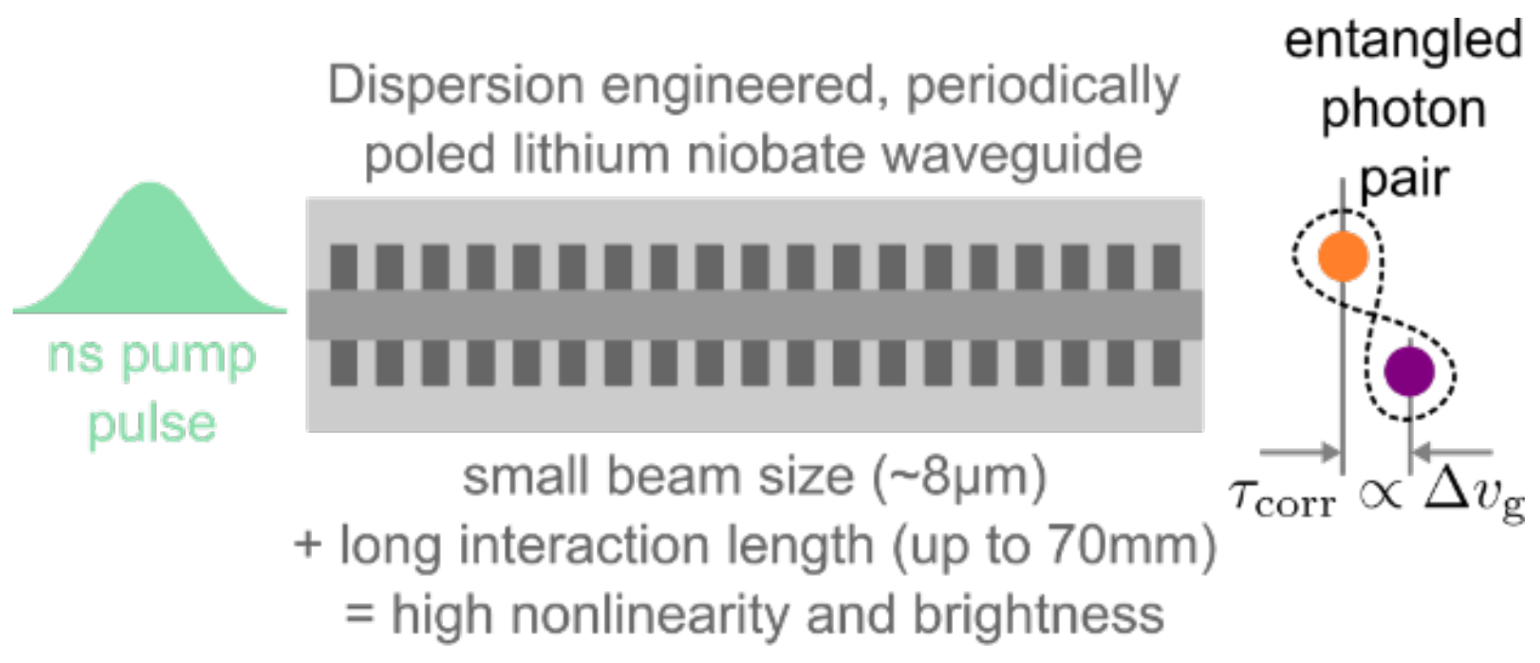


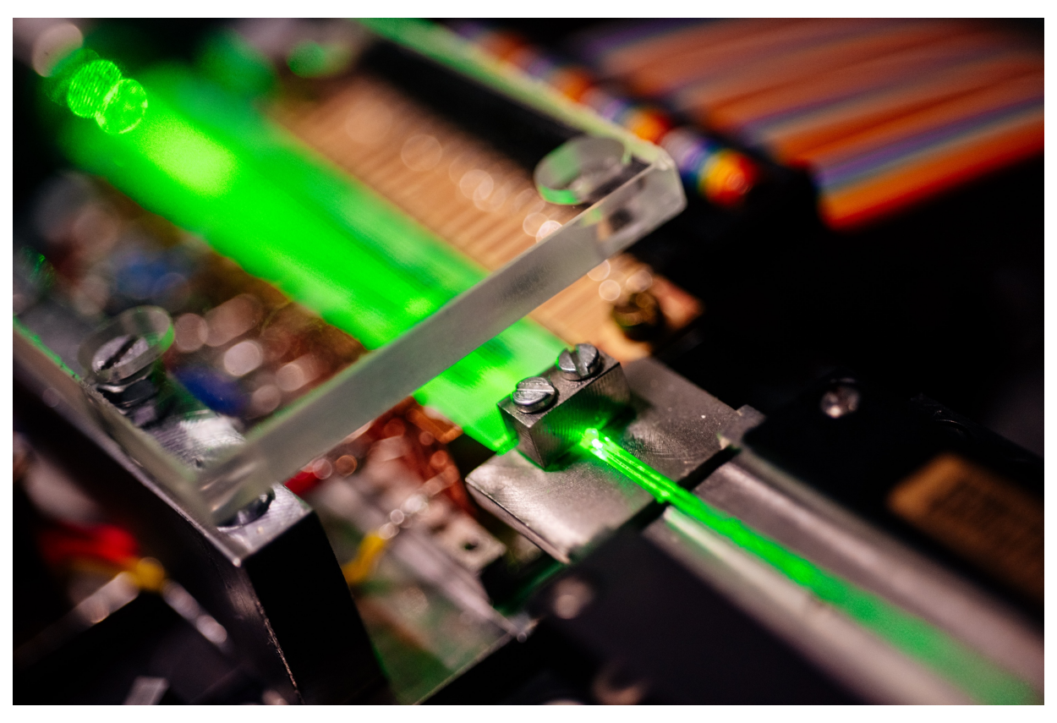
E²TPA Consortium

University of Geneva, Switzerland
Paderborn University, Germany
Nicolaus Copernicus University, Poland
University College London, United Kingdom
PicoQuant, Germany

PPLN Waveguide Technology

We will develop PPLN waveguide technologies for the relevant wavelength regimes, improving efficiencies - photon pair rates – for simpler/cheaper lasers and evaluating their performance for commercial fluorescence spectrometers.





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