INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

High harmonic generation: From attosecond science to quantum technologies

Paraskevas (Paris) Tzallas
FORTH, Crete, Greece: Research Director
ELI-ALPS, Szeged, Hungary: Scientific Advisor of SeSo Division
e-mail: ptzallas@iesl.forth.gr

ONASSIS Lectures, Heraklion, 3-7 July 2023

Part I: HHG and attosecond science

(High power attosecond light sources)

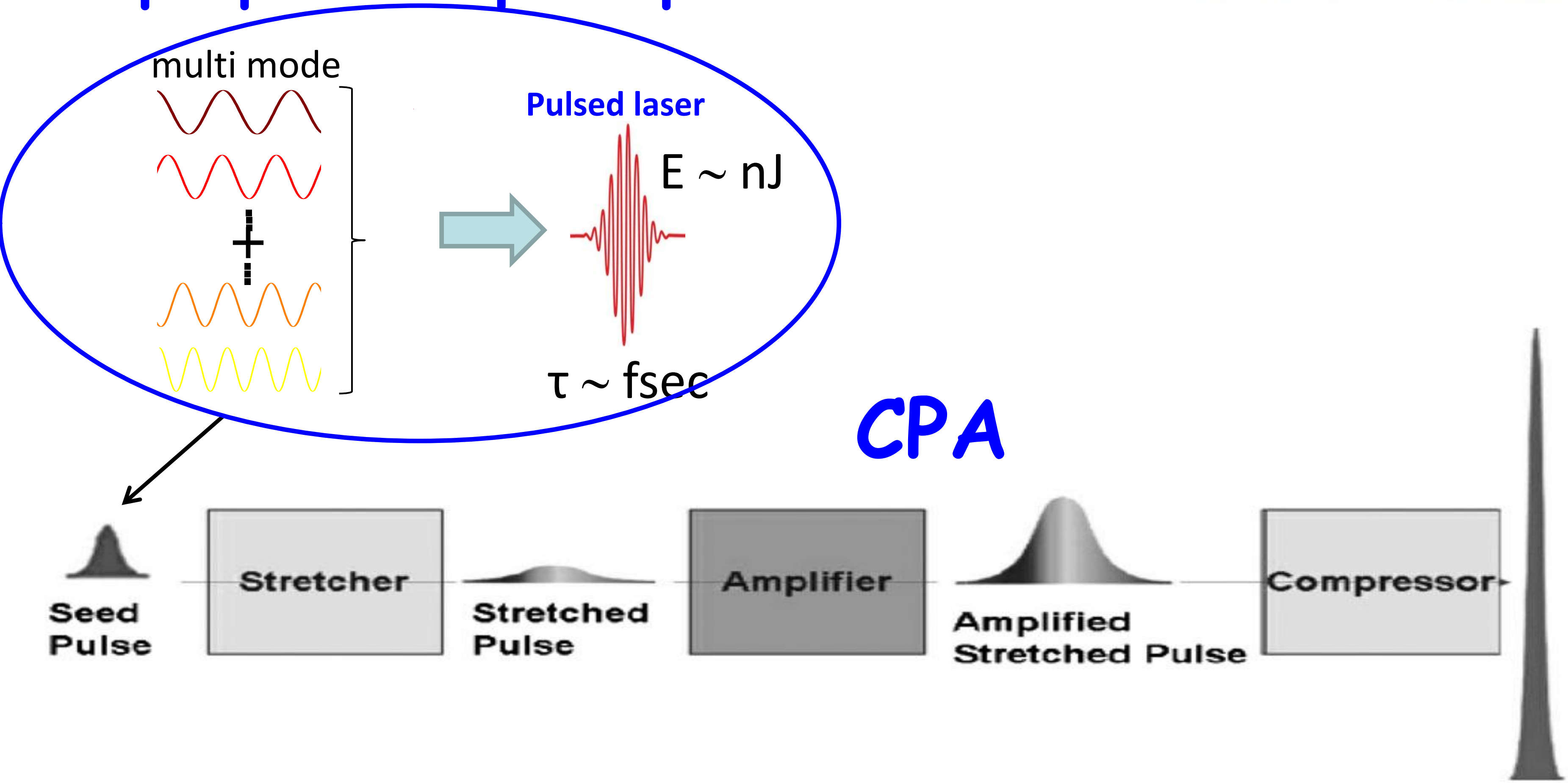
Part II: HHG and quantum technologies

(Generation of optical Schrödinger “cat” and entangled states)

FORTH
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

Superposition principle + CPA technique

Superposition principle

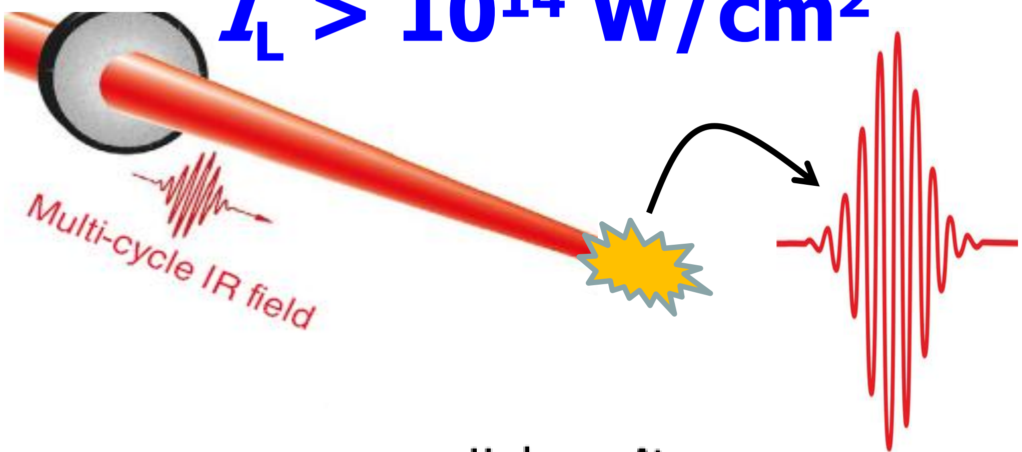


Laser systems in the TWatt, PWatt power level

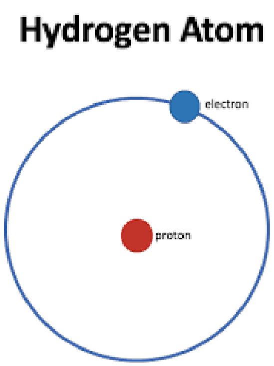
D. Strickland and G. Mourou, *Opt. Commun.* **56,** *219* (1985)
G. Mourou, *Rev. Mod. Phys.* **91**, *030501* (2019).
D. Strickland, *Rev. Mod. Phys.* **91,** *030502* (2019).

Strong laser-field physics

$I_L > 10^{14}$ W/cm^2



$E_L > 10^9$ V/cm



$E_{at} \sim 10^9$ V/cm

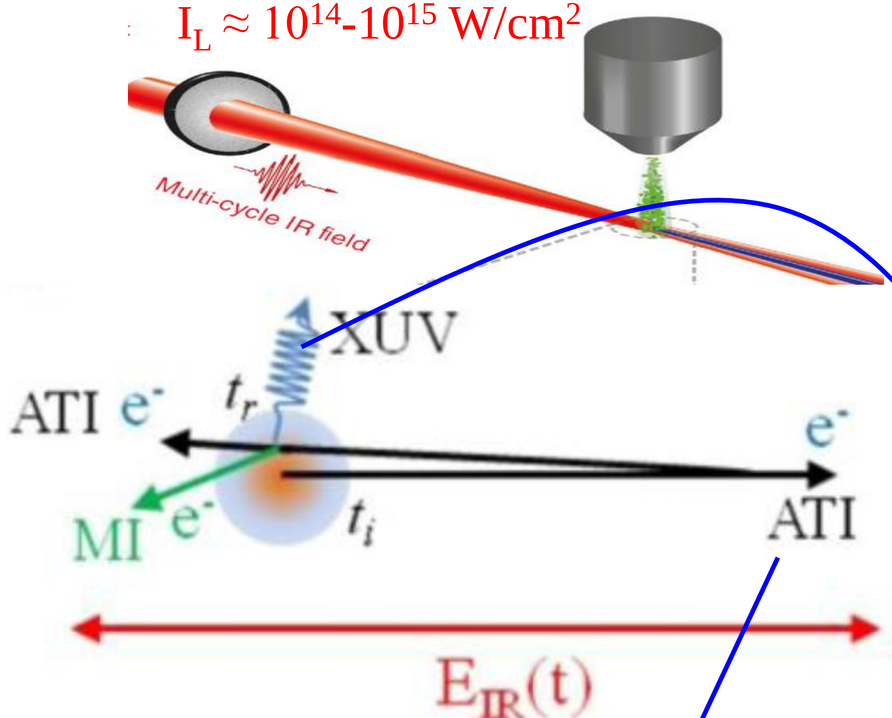
Strong laser-field physics:

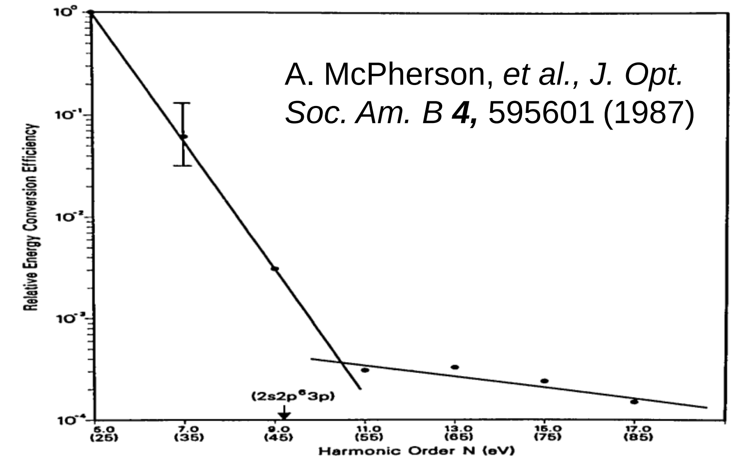
HHG, Attosecond Science, Laser-plasma physics, particle acceleration, relativistic optics e.t.c.

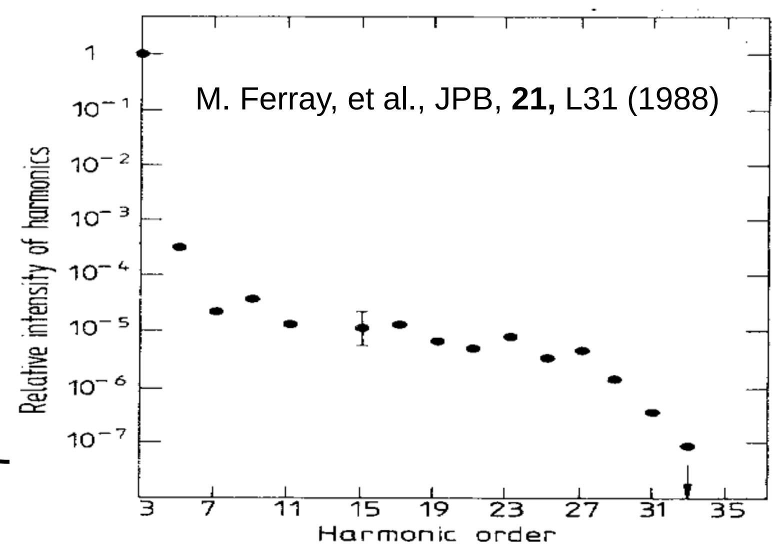
FORTH
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

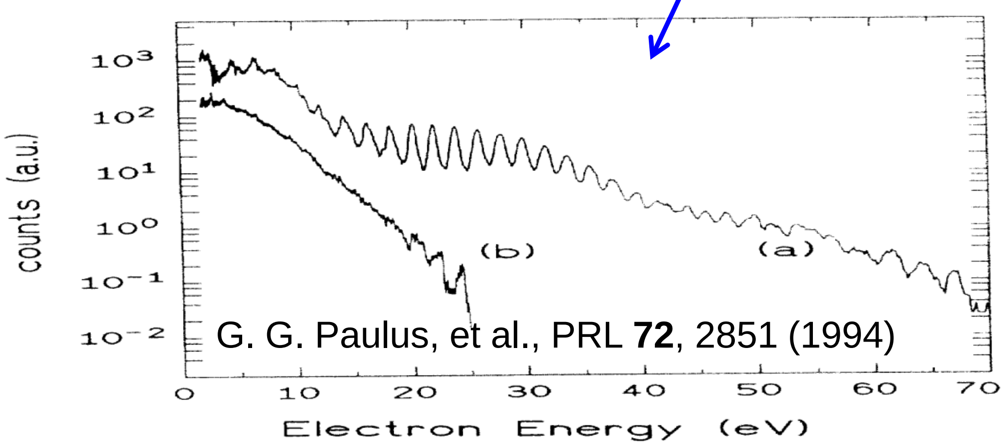
Strong field laser atom interaction and HHG

$I_L \approx 10^{14}$-10^{15} W/cm^2

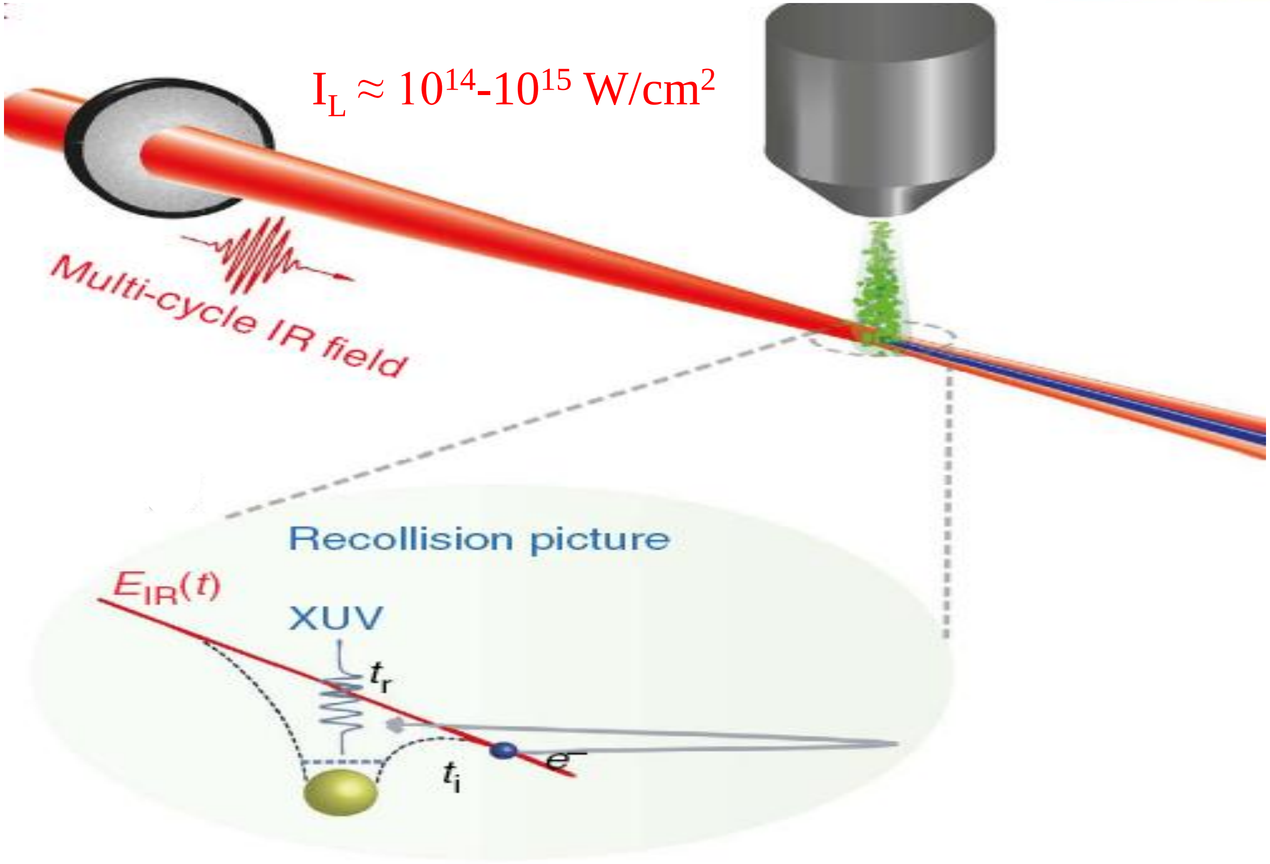








HHG: 3-step model

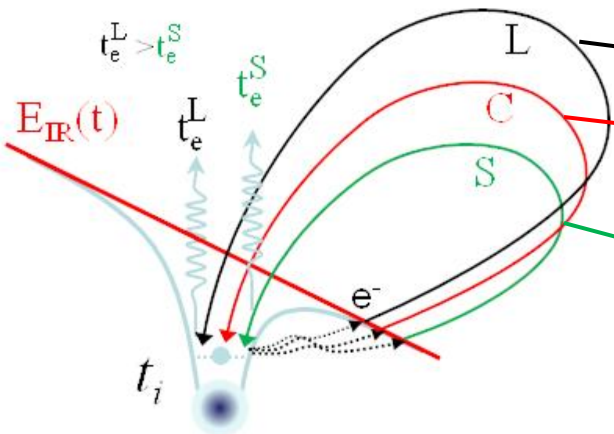


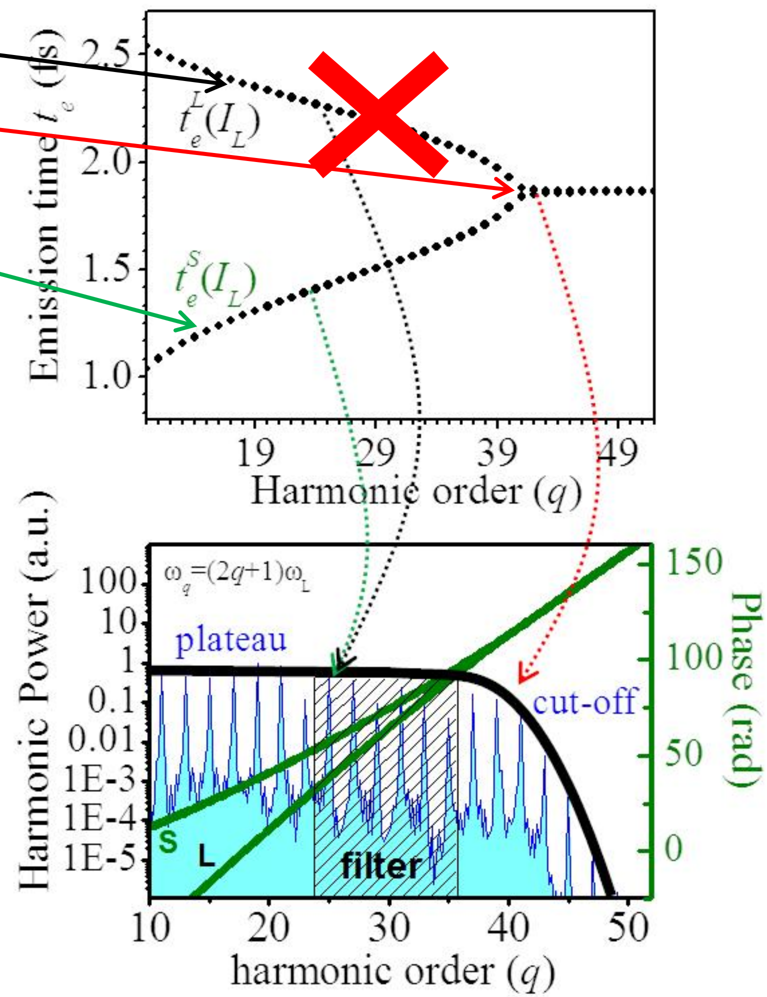
J. L. Krause, *et al.*, PRL **68**, 3535 (1992)

P. Corkum, PRL **71**, 1994 (1993)

M. Lewenstein, *et al.* PRA. **49**, 2117 (1994) (semi-classical approach → electron quantum paths)

HHG: 3-step model





$$E_{cut\text{-}off}=IP+3.17U_p$$

U_p (eV) $\approx 10^{-13}\, I(\text{W/cm}^2)\lambda^2(\mu\text{m})$

IP ~ 20 eV (atoms: He, Ne)

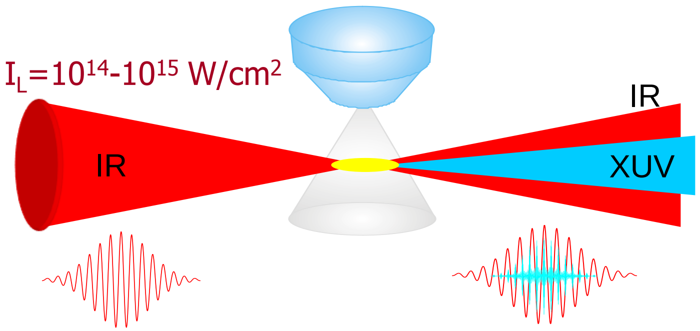
$I=2\text{x}10^{15}$ W/cm^2

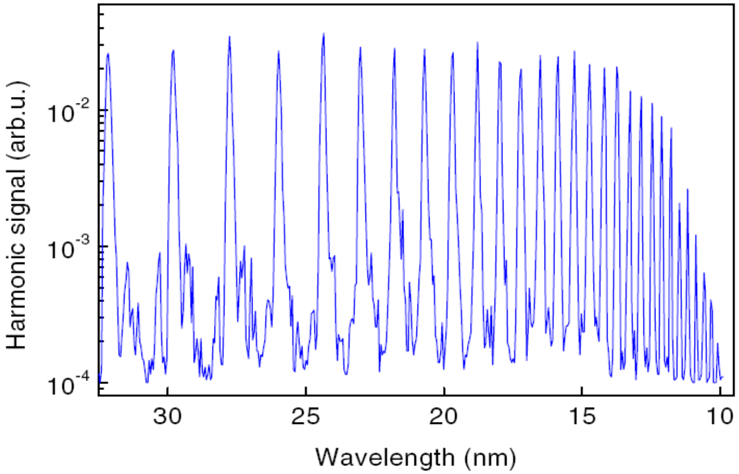
λ=800 nm

$E_{cut\text{-}off} \approx$ 450 eV (water window/soft-x-rays)

HHG in gas media

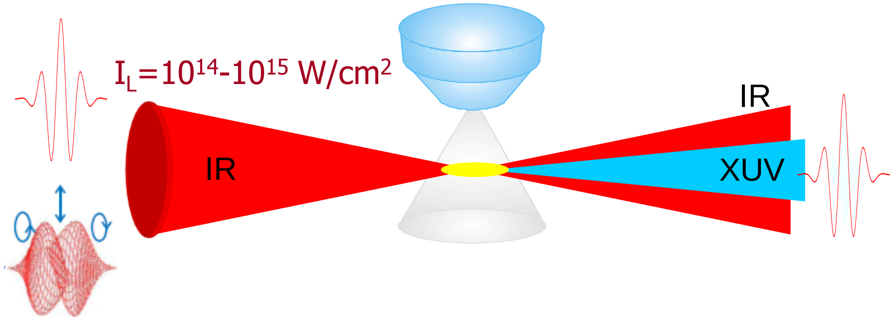
Generation of XUV comb

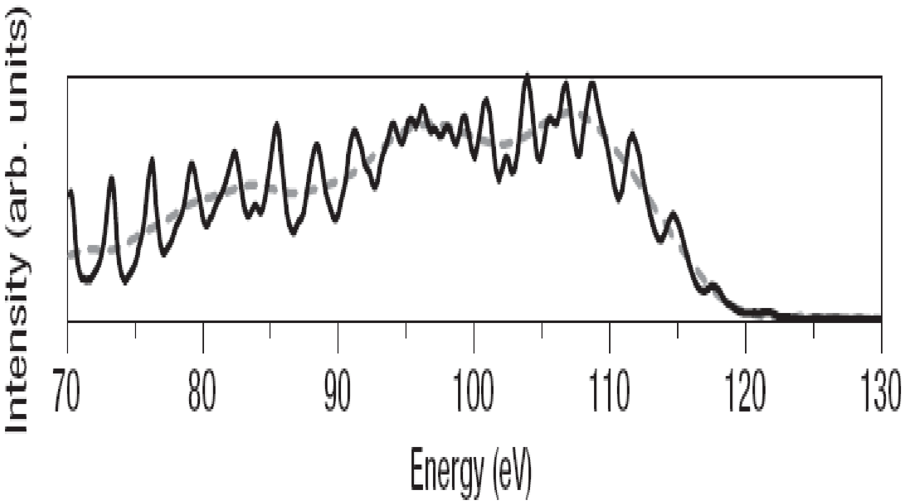




L. Le Deroff, *PhD thesis,* Paris 1999

Generation of XUV continuum





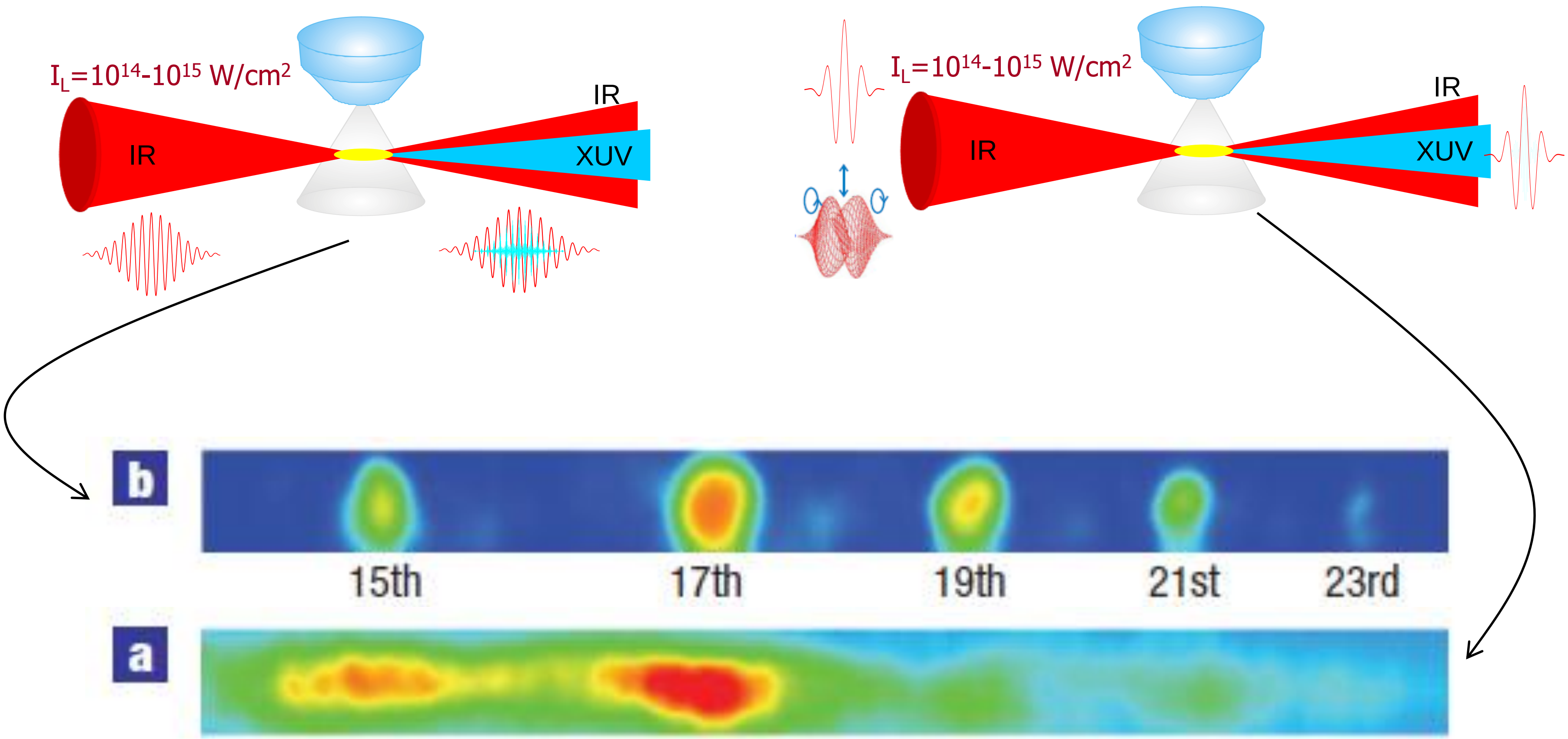
C.A. Haworth et al., *Nature Phys.* **3**, 52 (2007)

➢ **High harmonics are phase locked**

HHG in gas media

Generation of XUV comb

Generation of XUV continuum

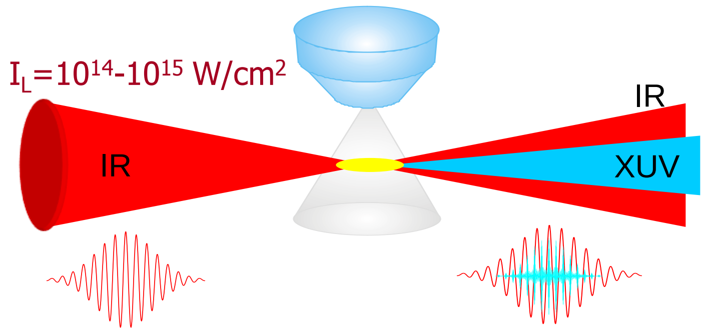


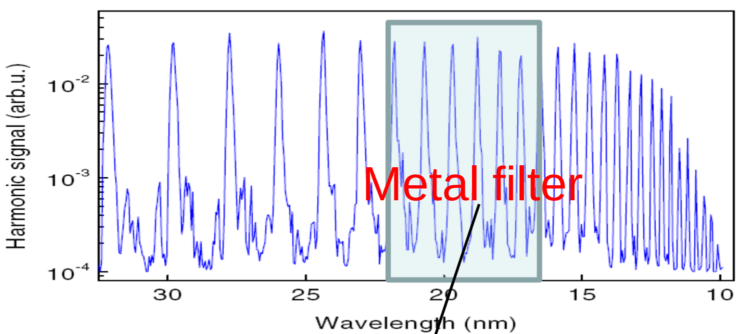
P. Tzallas et al., Nature Phys. **3**, 846, 2007

INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

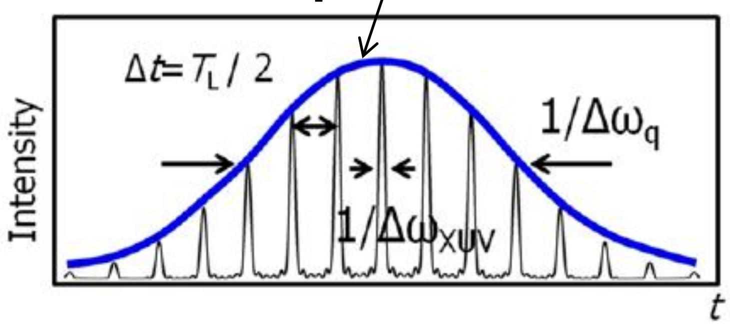
Attosecond pulses

Generation of XUV comb

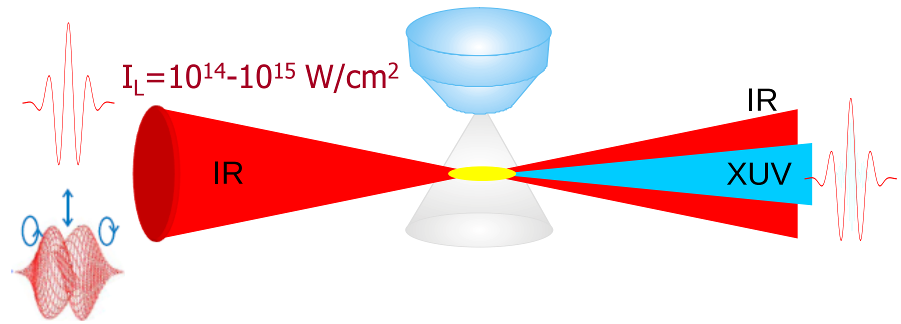


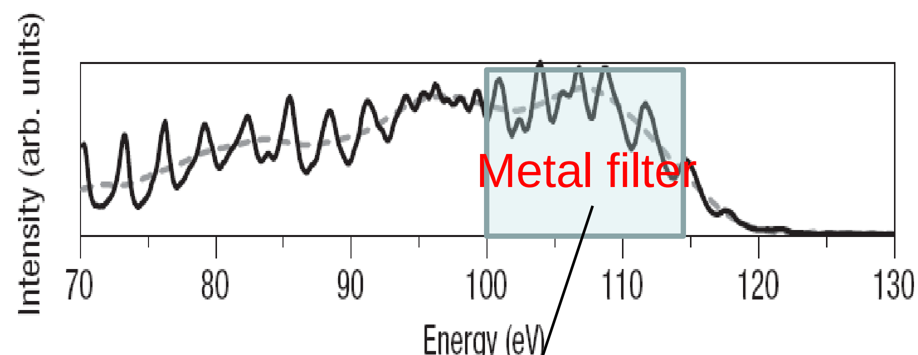


Asec pulse train

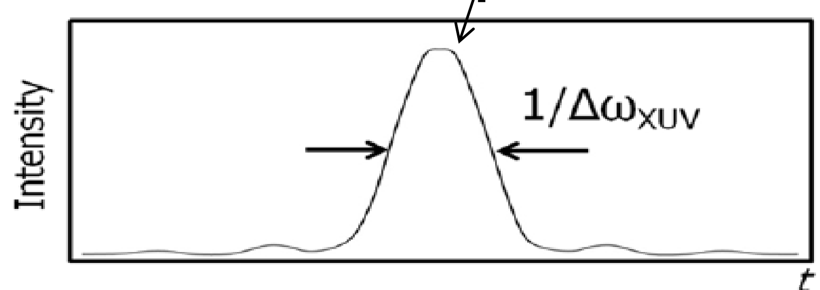


Generation of XUV continuum





Isolated *asec* pulse train



Temporal characterization of attosecond pulses

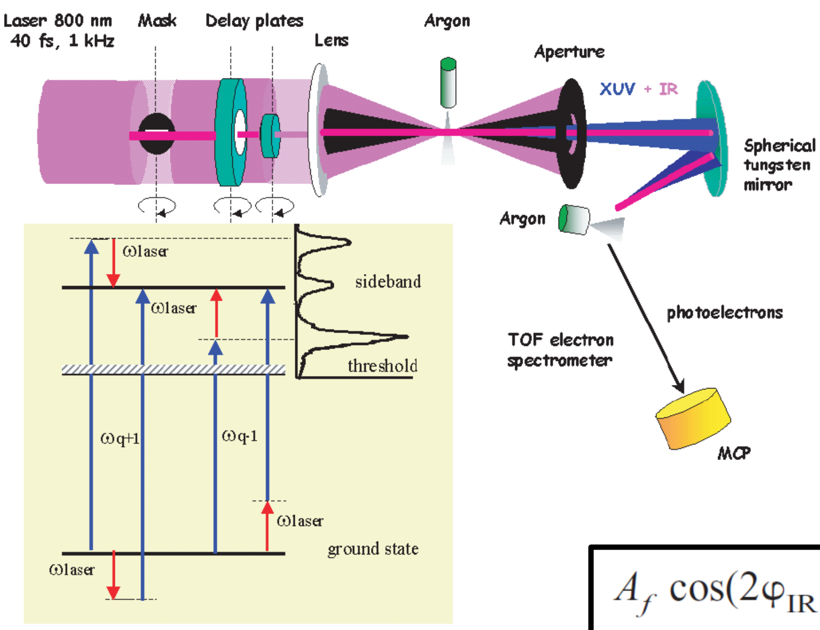
Tool box (development since 2001)

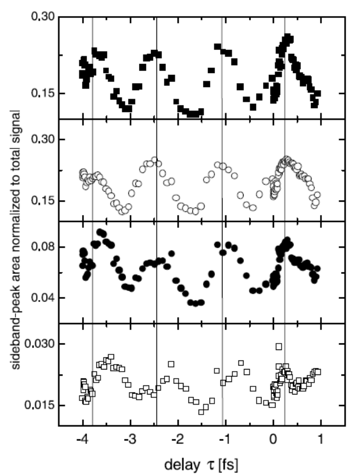
- **Cross-correlation (IR+XUV) methods (low energy atto pulses)**

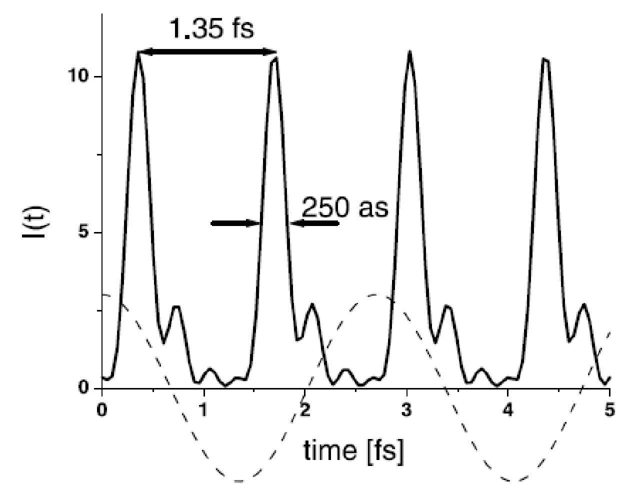
RABBITT

$$I_{\text{asec}} \sim 10^{8}\ \text{W/cm}^2$$

ATTO pulse train







$$A_f \cos(2\varphi_{\text{IR}} + \varphi_{q-1} - \varphi_{q+1} + \Delta\varphi^{f}_{\text{atomic}})$$

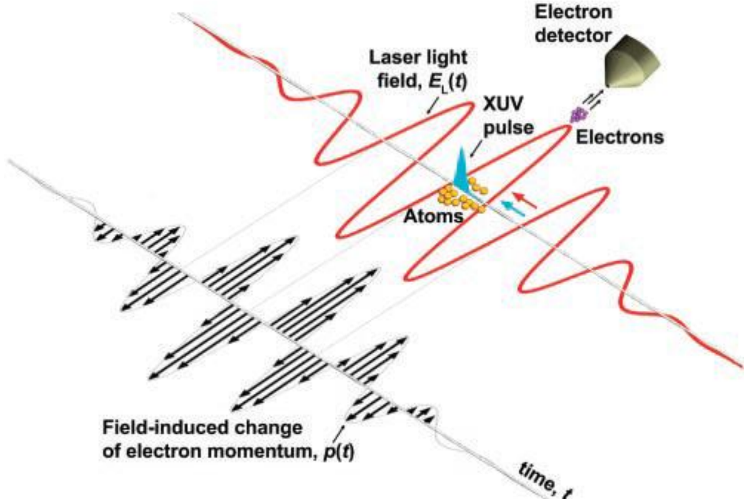
P. M. Paul et al., *Science* **292**, 1689 (2001)

Tool box (development since 2001)

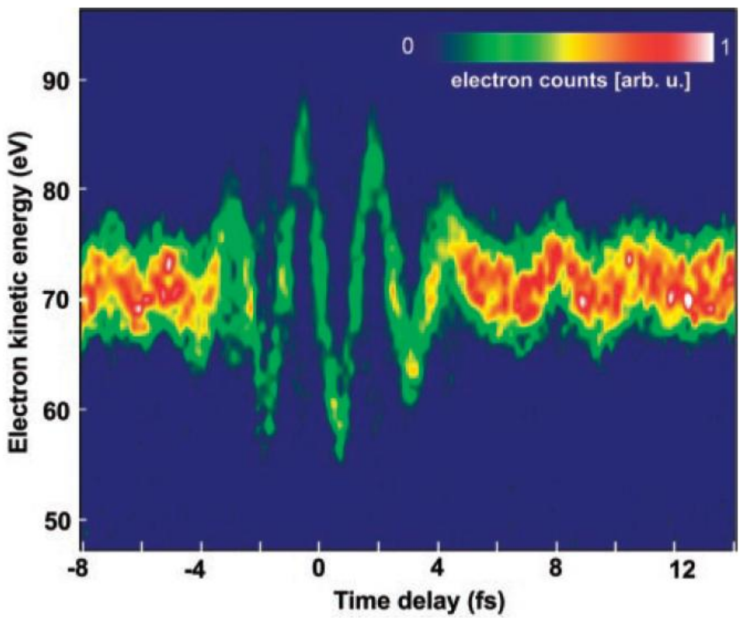
➢ **Cross-correlation (IR+XUV) methods (low energy atto pulses)**

$I_{asec} \sim 10^8\ \mathrm{W/cm^2}$

❑ **Streaking**



$$\Delta p(\vec{r},t) = e\int_t^{\infty} E_L(\vec{r},t')\mathrm{d}t' = eA_L(\vec{r},t)$$



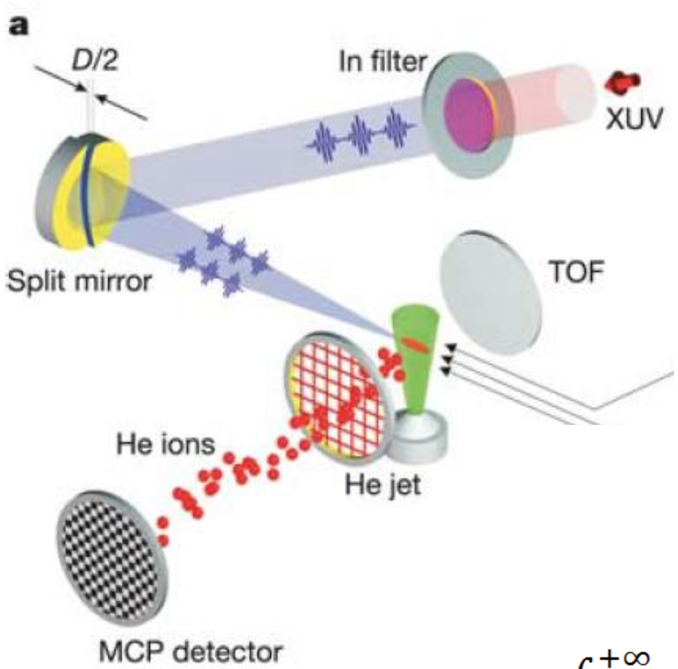
E. Goulielmakis et al., *Science* **305**, 1267 (2004)

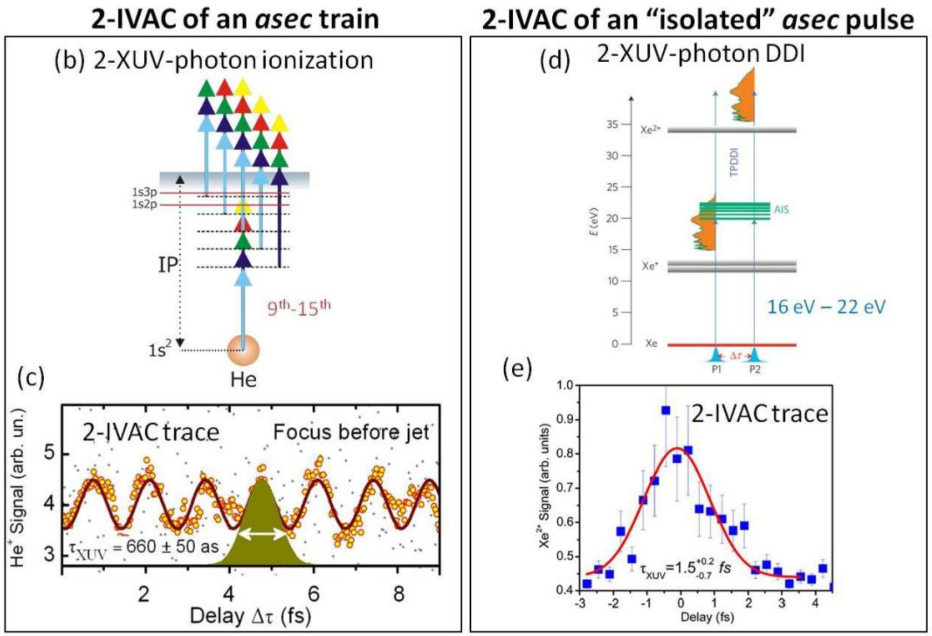
Temporal characterization of attosecond pulses

Tool box (development since 2001)

- 2nd order autocorrelation (XUV+XUV) (high energy atto pulses)

$I_{asec} > 10^{12}$ W/cm^2





$$S(\tau) \propto \int_{-\infty}^{+\infty} I_{XUV}(t,\tau) I_{XUV}(t) dt$$

P. Tzallas et al., *Nature* **426**, 267 (2003)

P. Tzallas et al., *Nature Phys.* **7**, 781, 2011

Tool box (development since 2001)

XUV FROG-type measurements

FROG-CRAB

D. Phase retrieval by omega oscillation filtering (PROOF)

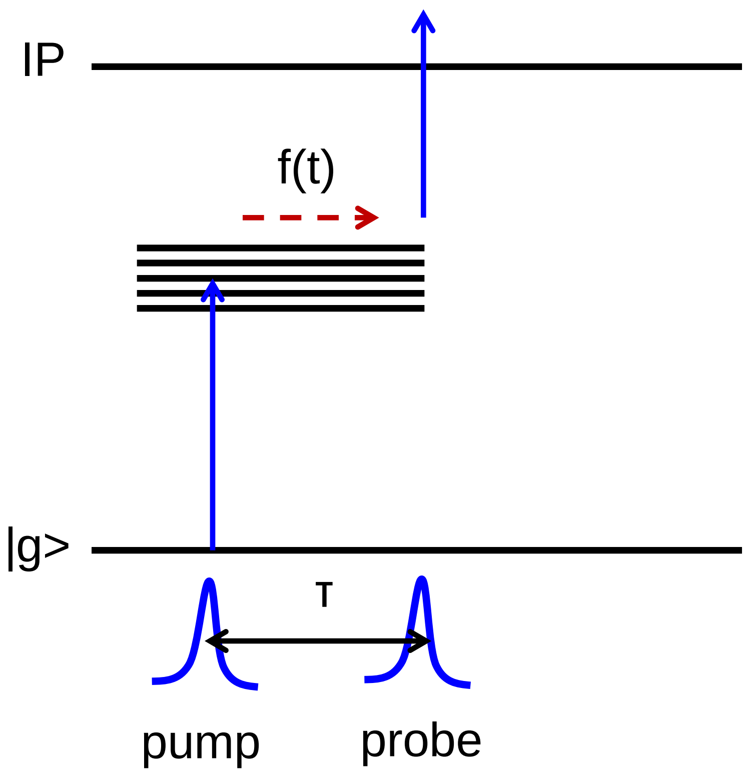
-
-
-

Review article on "Attosecond pulse metrology"

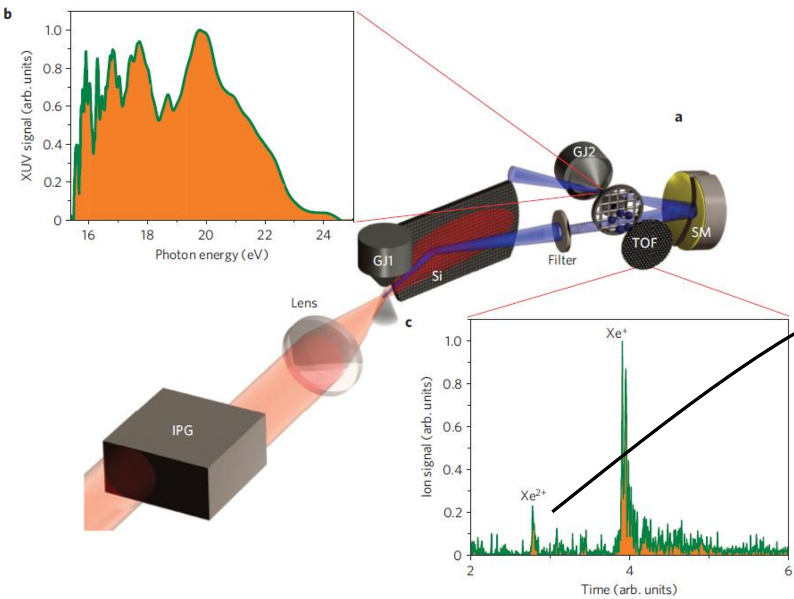
I. Orfanos *et al., APL Photonics* **4**, 080901 (2019)

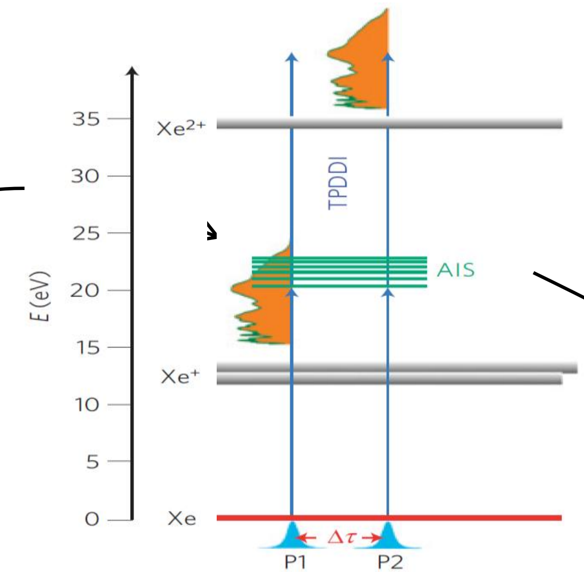
Ultrafast dynamics

Ion yield (e or ions) as a function of the delay τ

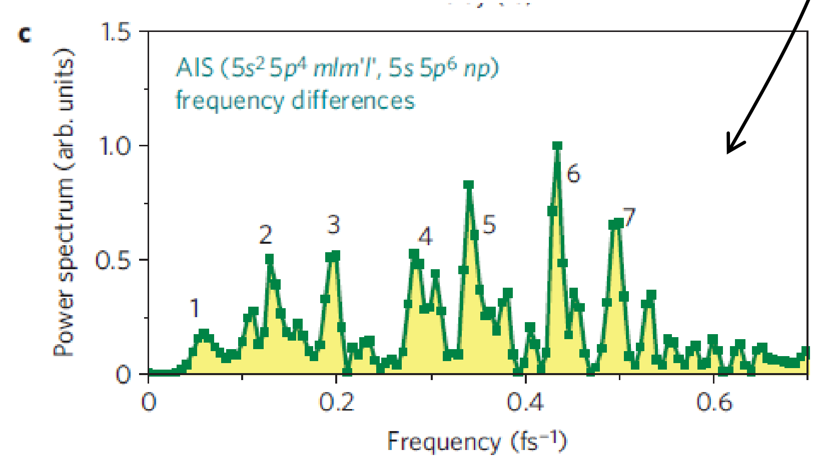


1-fs electron dynamics in atoms



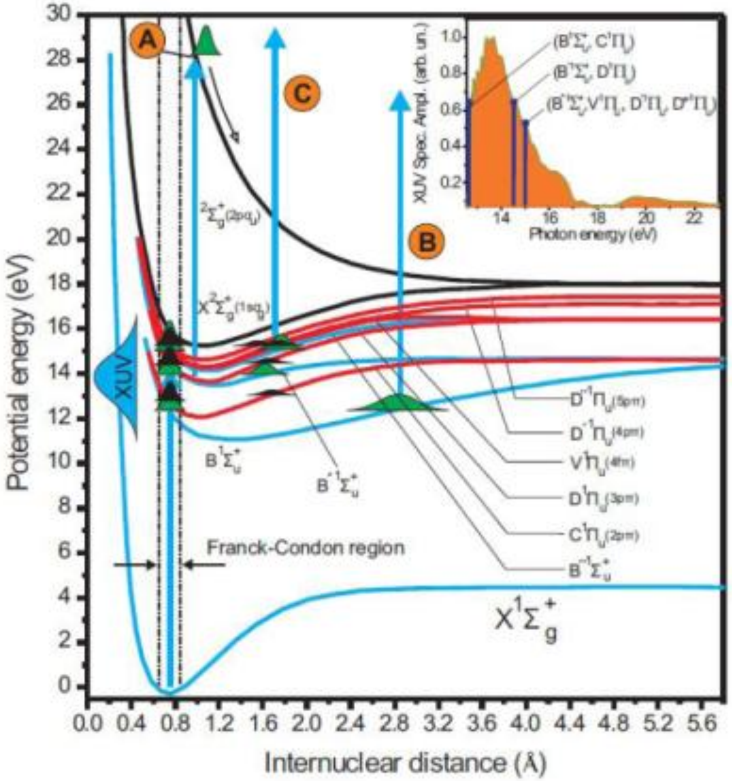


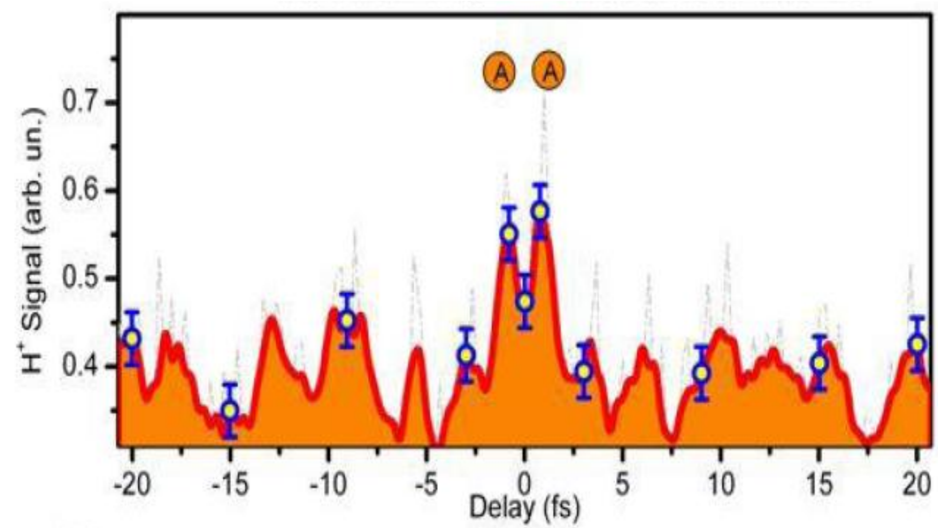
I_{asec}>10^{12} W/cm^2

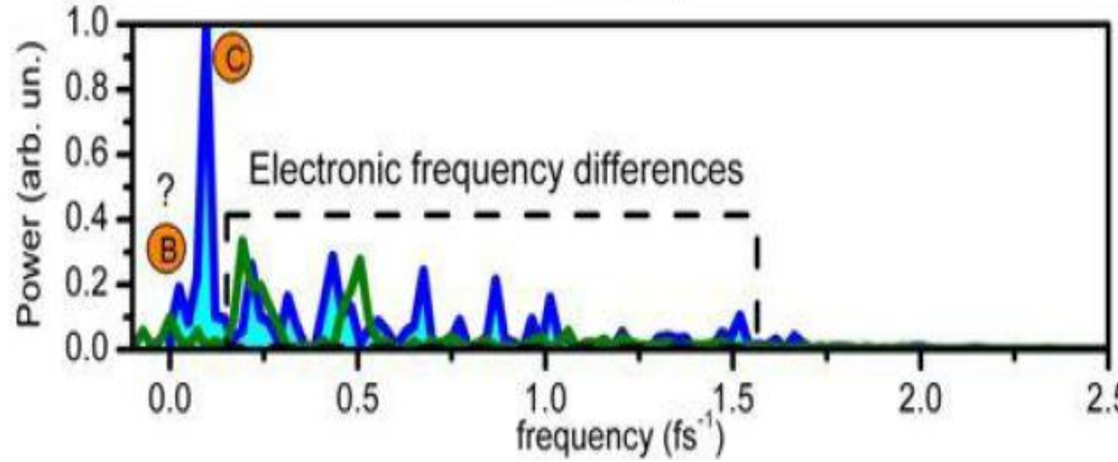


P. Tzallas et al., Nature Phys. **7**, 781, 2011

1-fs nuclear dynamics in molecules







P. Carpegianni et al., PRA, **89**, 023420 (2014)

High power attosecond source

$I_{IR}=E/\tau A=ct\approx 10^{14}\ W/cm^2$

$A=\pi R^2$

$Y_{atto}\propto A$

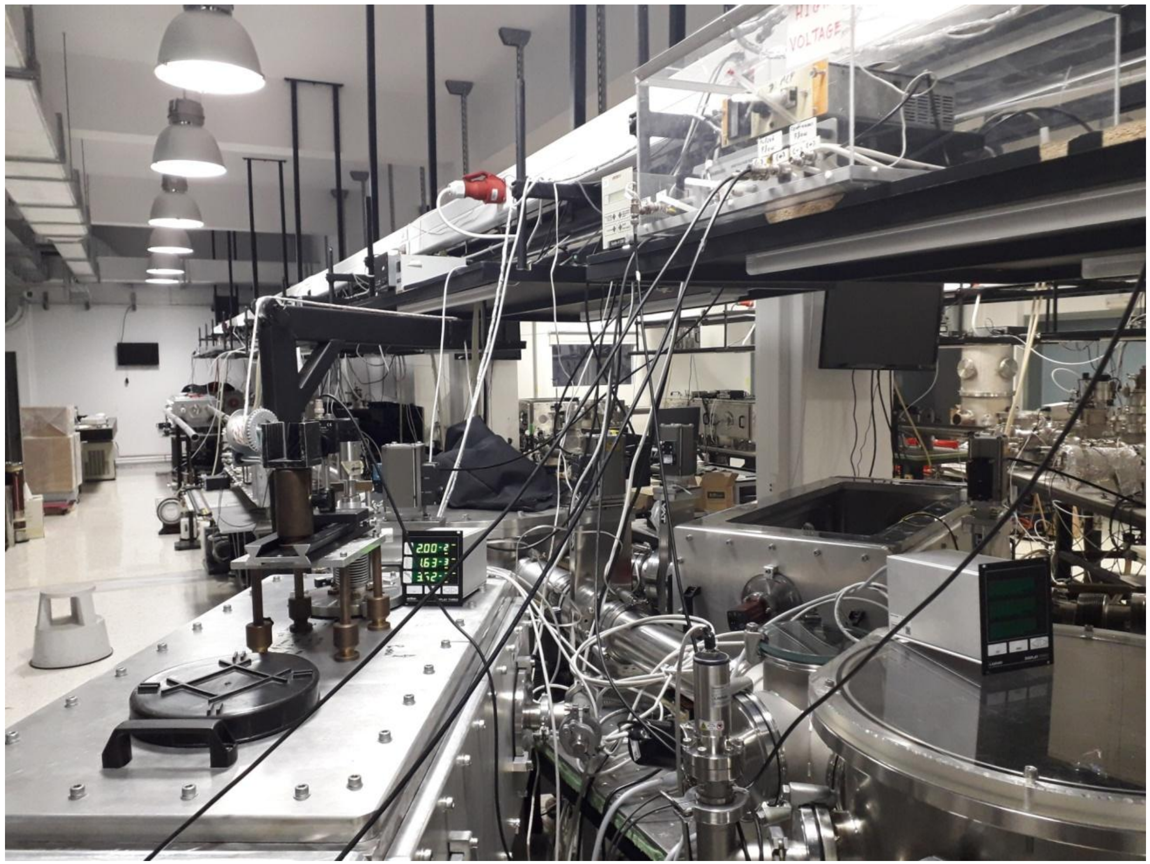
$Y_{atto}\uparrow$ E, A $\uparrow$ $I_{IR}=ct$

High power IR fs laser and loose focusing geometries (long beam lines)

The world's highest power fs-asec XUV source

20-GWatt attosecond beam line at FORTH-IESL

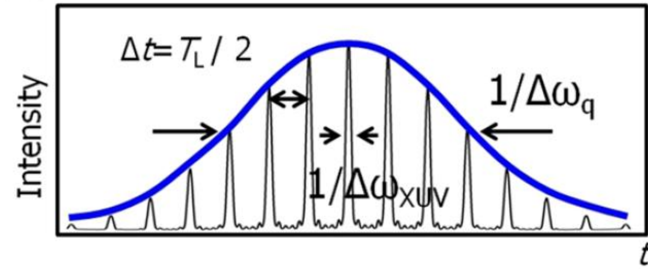
A. Nayak et al., *Phys. Rev. A* 98, 023426 (2018)



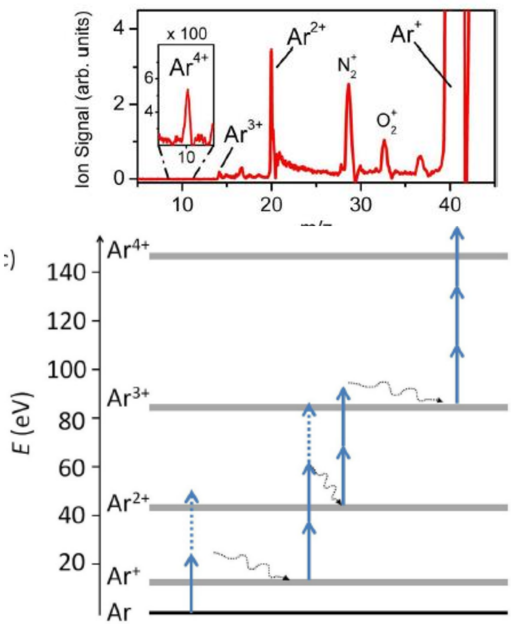
$$I_{asec} > 10^{15}\ \mathrm{W/cm^2}$$

Recent achievements

Using 10 fs XUV pulses (asec train):

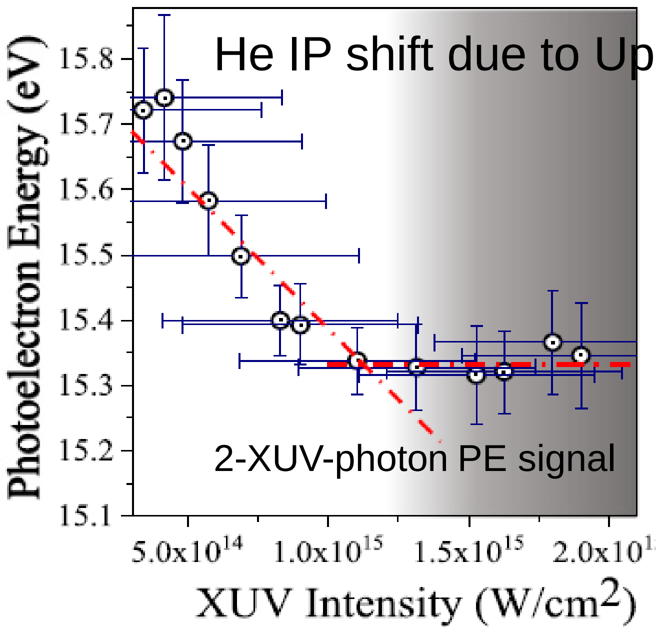


Multiphoton multiple ionization in XUV



A. Nayak et al., *PRA* **98**, 023426 (2018)
I. Makos *et al., Sci. Rep.* **10**, 3759 (2022)

Strong field effects in XUV

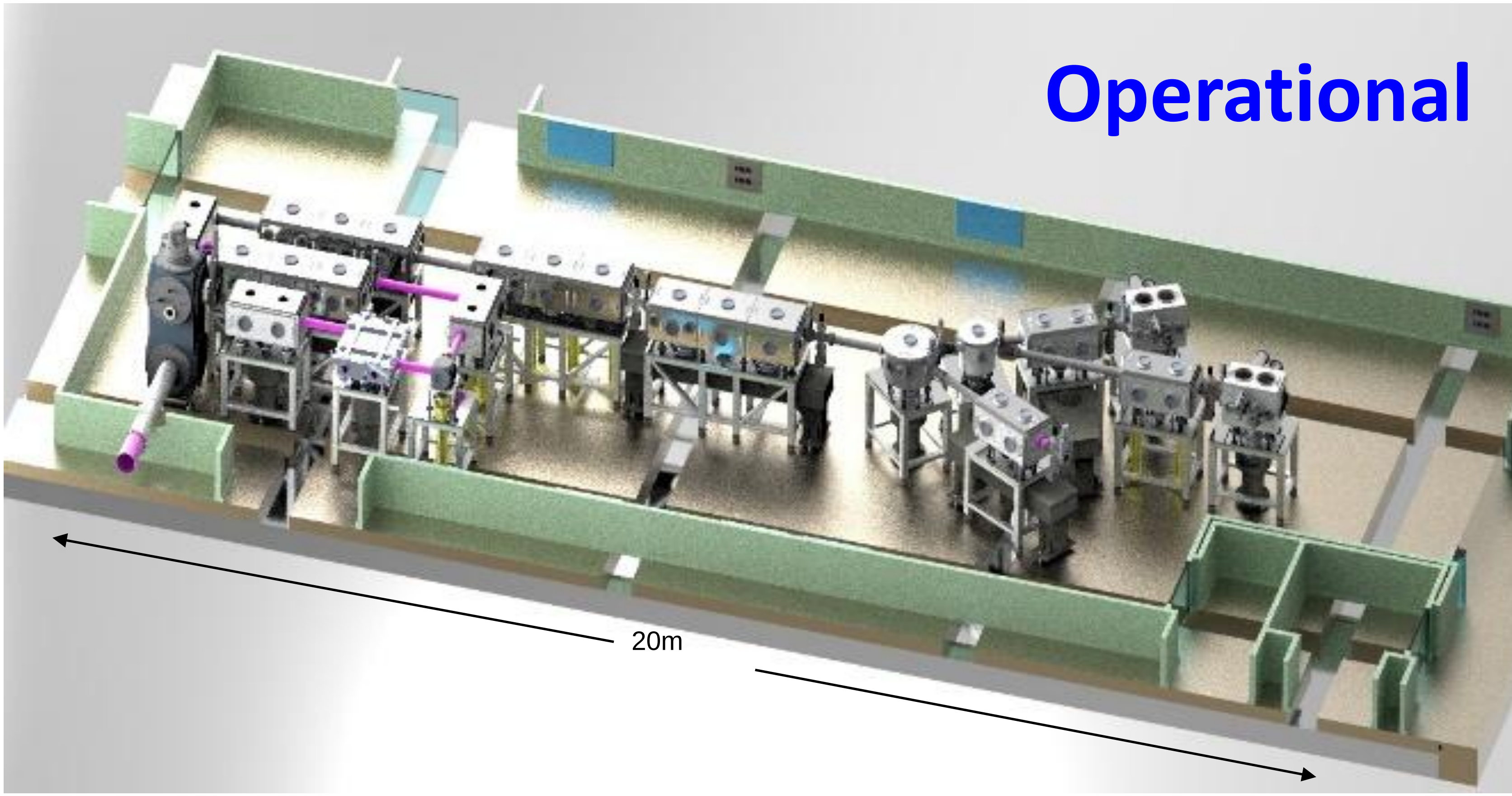


I. Orfanos et al., *J. Phys. B*, **54**, 084002 (2021)

SYLOS beam line at ELI-ALPS: Developed by FORTH-IESL

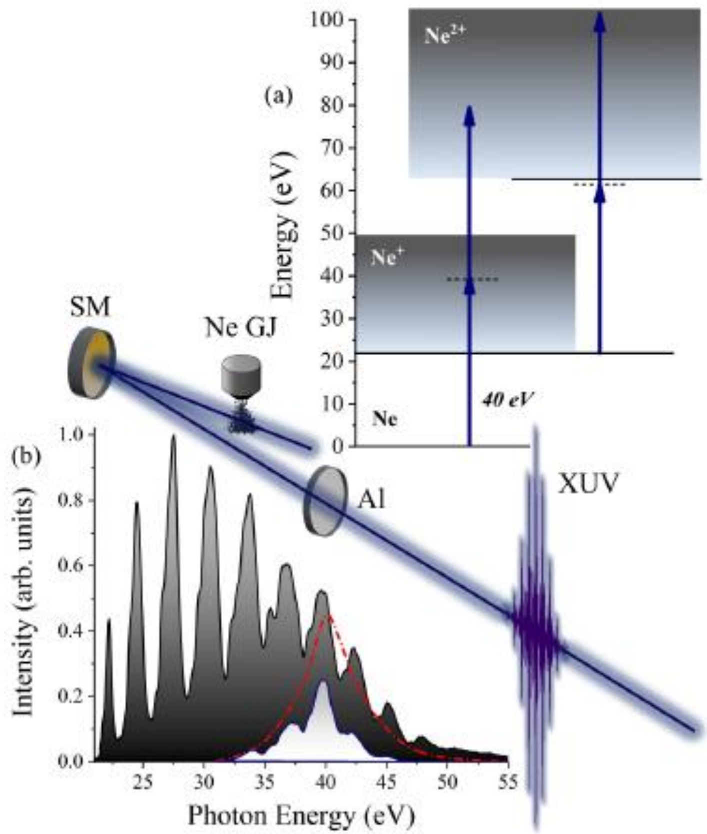
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

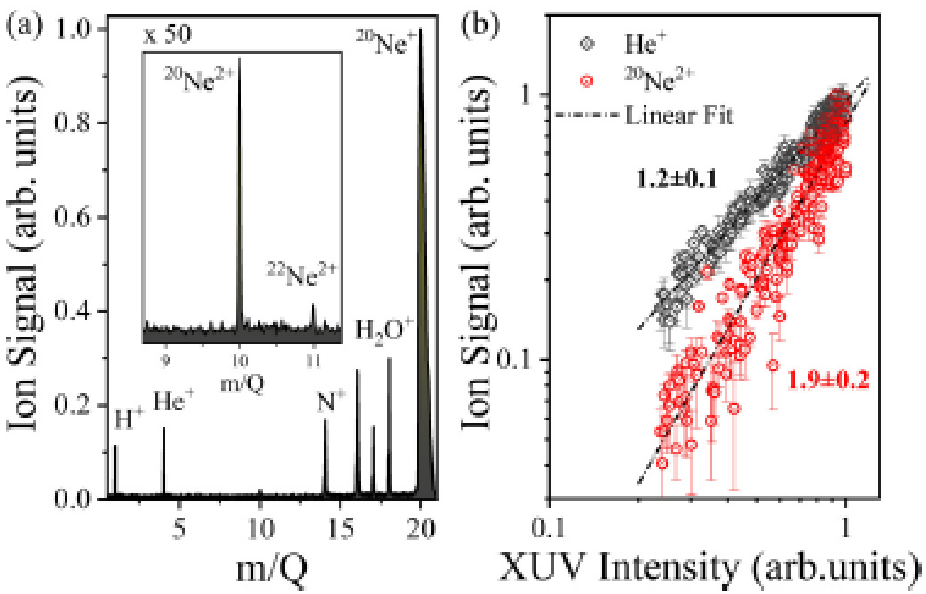
Intense asec pulses at 1kHz rep. rate



Available for users

SYLOS beam line at ELI-ALPS: Developed by FORTH-IESL





I. Orfanos et al., PRA **106**, 043117 (2022)

Acknowledgments:

FORTH
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

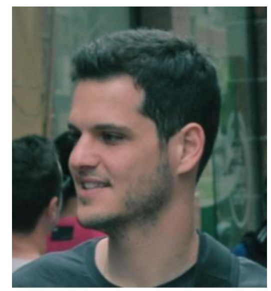
Former members

P. Tzallas

D. Charalambidis

E. Skantzakis

E. Vassakis



I. Orfanos

...

International collaborators

eli attosecond

DCU
Dublin city university

MPQ
Max Planck Institute
for Quantum Optic

...and many others

PART II:
Generation of optical Schrödinger ”cat” and entangled states using intense laser-atom interactions

Schrödinger's "cat"

E. Schrödinger, *Naturwissenschaften*, **23**, 807-812 (1935)

Cat state: $|\Psi\rangle = \frac{1}{\sqrt{2}}(|dead\rangle + |alive\rangle)$

Entanglement: $|\Psi\rangle = \frac{1}{\sqrt{2}}(|decay\rangle|dead\rangle + |no\ decay\rangle|alive\rangle)$

Why is so hard to prepare and observe this behavior?

Schrödinger "cat" state

The reason is that for macroscopic systems, quantum superpositions rapidly decohere into statistical mixtures due to interactions with the environment.

$$\tau_d \approx \tau_r \left(\frac{\lambda_{dB}}{\Delta x}\right)^2$$

W. H. Zurek *Phys. Today* **36** (1991)

$\lambda_{dB} = \hbar/\sqrt{2mk_BT}$ → De Broglie wavelength

m → Mass of the particle

k_B → Boltzmann constant

T → Temperature

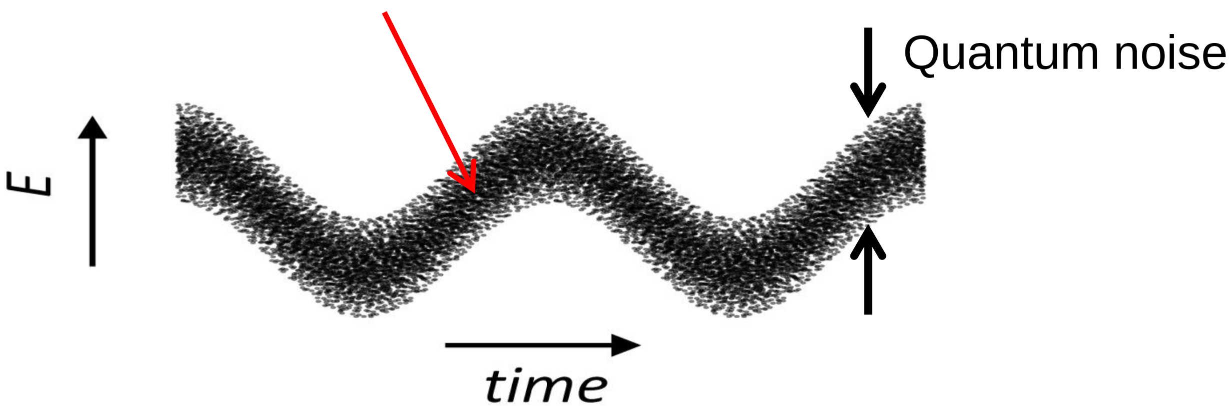
1) Macroscopic objects:
m=100kg, T=300K, Δx=1cm, τ_r=100 years

⇨ $\tau_d \sim 10^{-36}$ sec

Schrödinger "cat" state

To become feasible, Schrodinger Gedankenexperiment in macroscopic scale we should replace the cat with a physical system with its own classical states. This can be done using the coherent light states |a>, although purely quantum, the considered as classical because they discribe a classically EM field (as the laser field)

$$\langle a|\hat{E}|\alpha\rangle = |a|\cos(\omega t + \varphi)$$



Schrödinger "cat" state using coherent light states

Schrödinger "cat" state: $|\Psi\rangle = \frac{1}{\sqrt{2}}(|dead\rangle + |alive\rangle)$

⬇

Optical Schrödinger "cat" state:

$$|\Psi\rangle = A\,(|\alpha\rangle \pm |-\alpha\rangle)$$

or in a more general form

$$|\Psi\rangle = A|a\rangle \pm B|b\rangle$$

Optical cat states is one of the main recourses for investigations in Quantum technology:

- **Quantum information processing,**
- **Quantum metrology/sensing,**
- **Fundamental tests of quantum theory**

.

.

.

A. Acín et al., *New J. Phys.* ***20,*** 080201 (2018)***.***
I. A. Wamsley *Science* 348, **525** (2015)
I. H. Deutsch *PRX Quantum* **1**, 020101 (2020)
A. Gilchrist *et al., J. Opt. B: Quantum Semiclass. Opt.* **6**, S828 (2004)
V. Giovanneti et al., Nat. Photon. **5**, 222 (2011)

Why?????

Optical Schrödinger "cat" state

A. Gilchrist et al., *J. Opt. B: Quantum Semiclass. Opt.* **6**, S828 (2004)

1. Introduction

Quantum optics has played a major role in the testing of fundamental properties of quantum mechanics and more recently in implementing simple quantum information protocols [1, 2]. This has been made possible because photons are easily produced and manipulated. This is especially true as the electromagnetic environment at optical frequencies can be regarded as a vacuum and is relatively decoherence free.

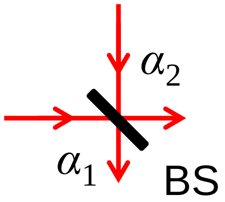
Toolbox for applications in quantum technology

Passive linear optical elements

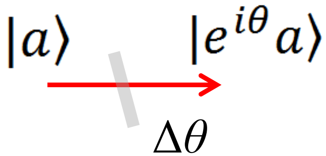
1) Coherent light states: $|\alpha\rangle$ (Many times is used to displace a state)

2) Optical cat state sources: $|\Psi\rangle = A\,(|\alpha\rangle \pm |-\alpha\rangle)$

3) Beam splitters: $\hat{B}(T) = e^{\arccos(\sqrt{T})(\hat{a}_1\hat{a}_2^\dagger - \hat{a}_1^\dagger\hat{a}_2)}$

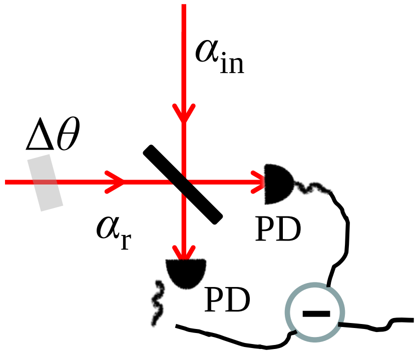


4) Phase shifters: $\hat{P}(\theta) = \exp\,[i\theta\hat{a}^\dagger\hat{a}]$



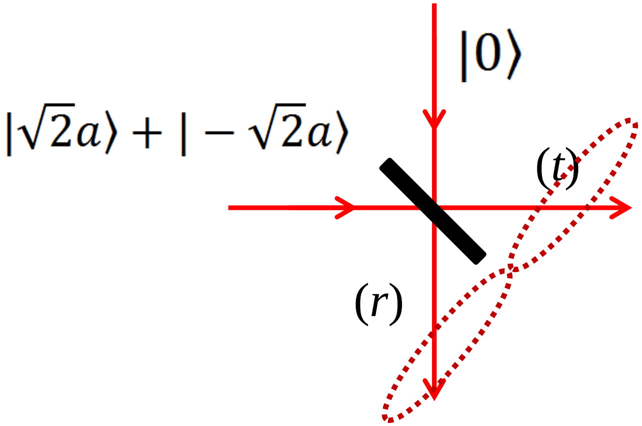
5) Photon counters:

6) Homodyne detection units:



Massively entangled Bell states

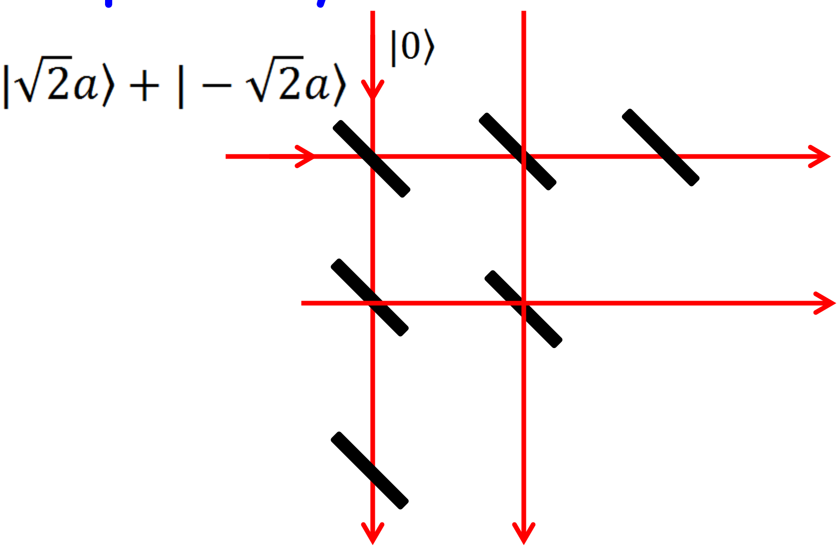
Optical cat state on a beam splitter



Entangled state

$$|\Psi\rangle = B(|a\rangle_r |a\rangle_t + |-a\rangle_r |-a\rangle_t)$$

N port symmetric beam splitter



Massively entangled states

$$|a, a, \dots, a\rangle + |-a, -a, \dots, -a\rangle$$

Quantum light state characterization

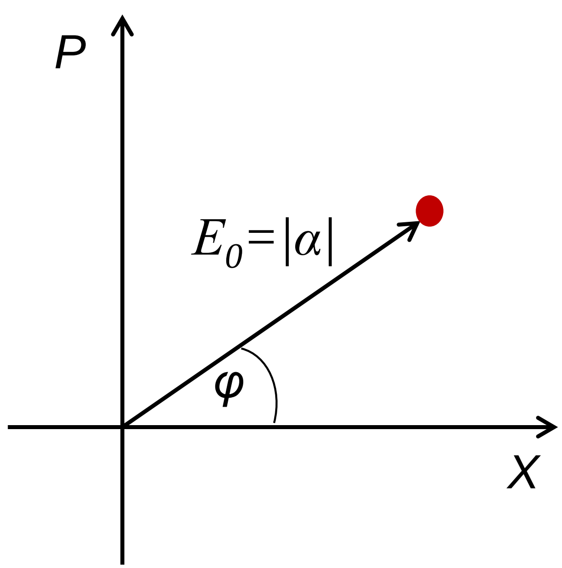
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

Field distribution: Phase space representation

Classical field

$$E(t) = E_0 \cos(\omega t + \varphi)$$
$$= \frac{1}{2}\left(a e^{i\omega t} + a^* e^{-i\omega t}\right)$$
$$= X\cos(\omega t) + P\sin(\omega t)$$

$$a = E_0 e^{i\varphi} = X + iP$$
$$X = E_0 \cos(\varphi) = Re[a] = \frac{1}{2}(a + a^*)$$
$$P = E_0 \sin(\varphi) = Im[a] = \frac{1}{2i}(a - a^*)$$

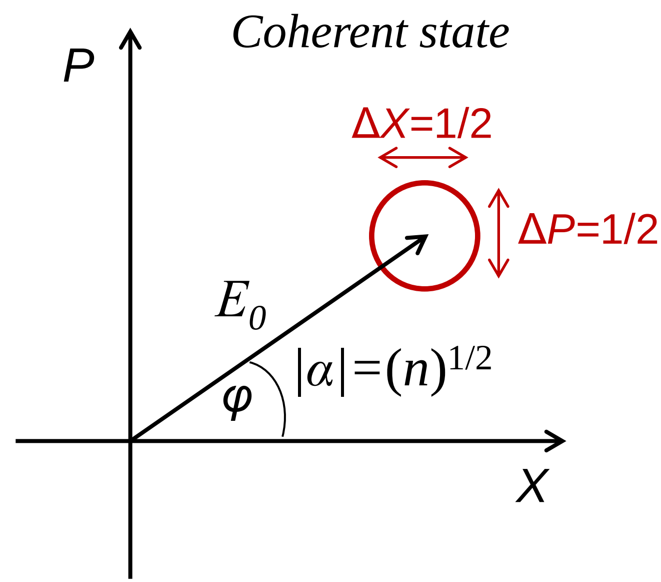


Quantum field states

$$\hat{E}(t) = \hat{X}\cos(\omega t) + \hat{P}\sin(\omega t)$$

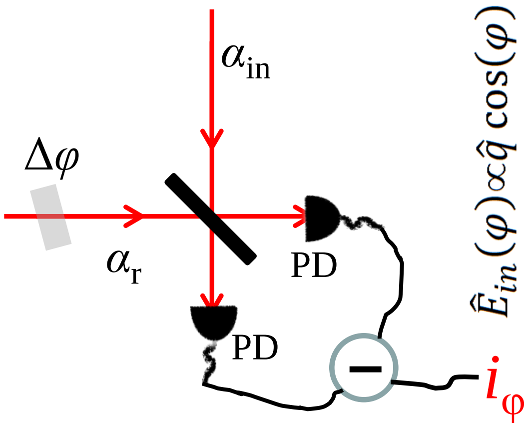
Quantum harm. osc.

$$\hat{X} = \frac{1}{2}\left(\hat{a} + \hat{a}^\dagger\right) \rightarrow \text{position}$$
$$\hat{P} = \frac{1}{2i}\left(\hat{a} - \hat{a}^\dagger\right) \rightarrow \text{momentum}$$
$$\hat{a} = \hat{X} + i\hat{P} \quad \hat{a}^\dagger = \hat{X} - i\hat{P}$$
$$\left[\hat{X}, \hat{P}\right] = \mathrm{i}/2 \quad \Delta X \Delta P \geq 1/4$$



Quantum state characterization

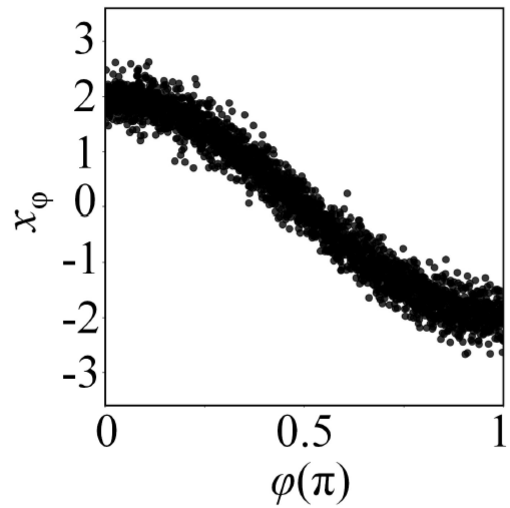
Quantum Tomography

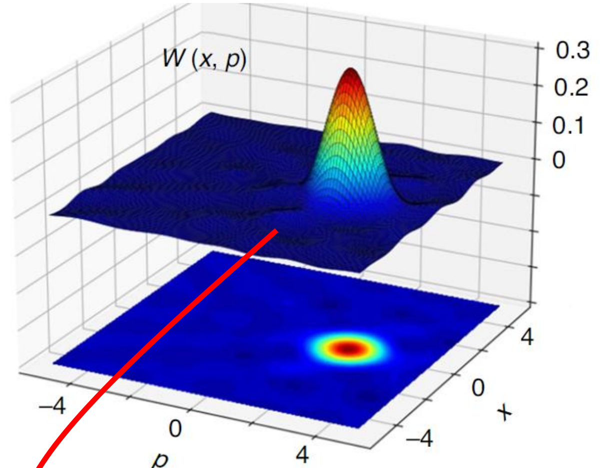


$a_{in} << \alpha_r$

$$\hat{E}_{in}(\varphi) \propto \hat{q}\cos(\varphi) + \hat{p}\sin(\varphi)$$

Wigner function of a coherent light state



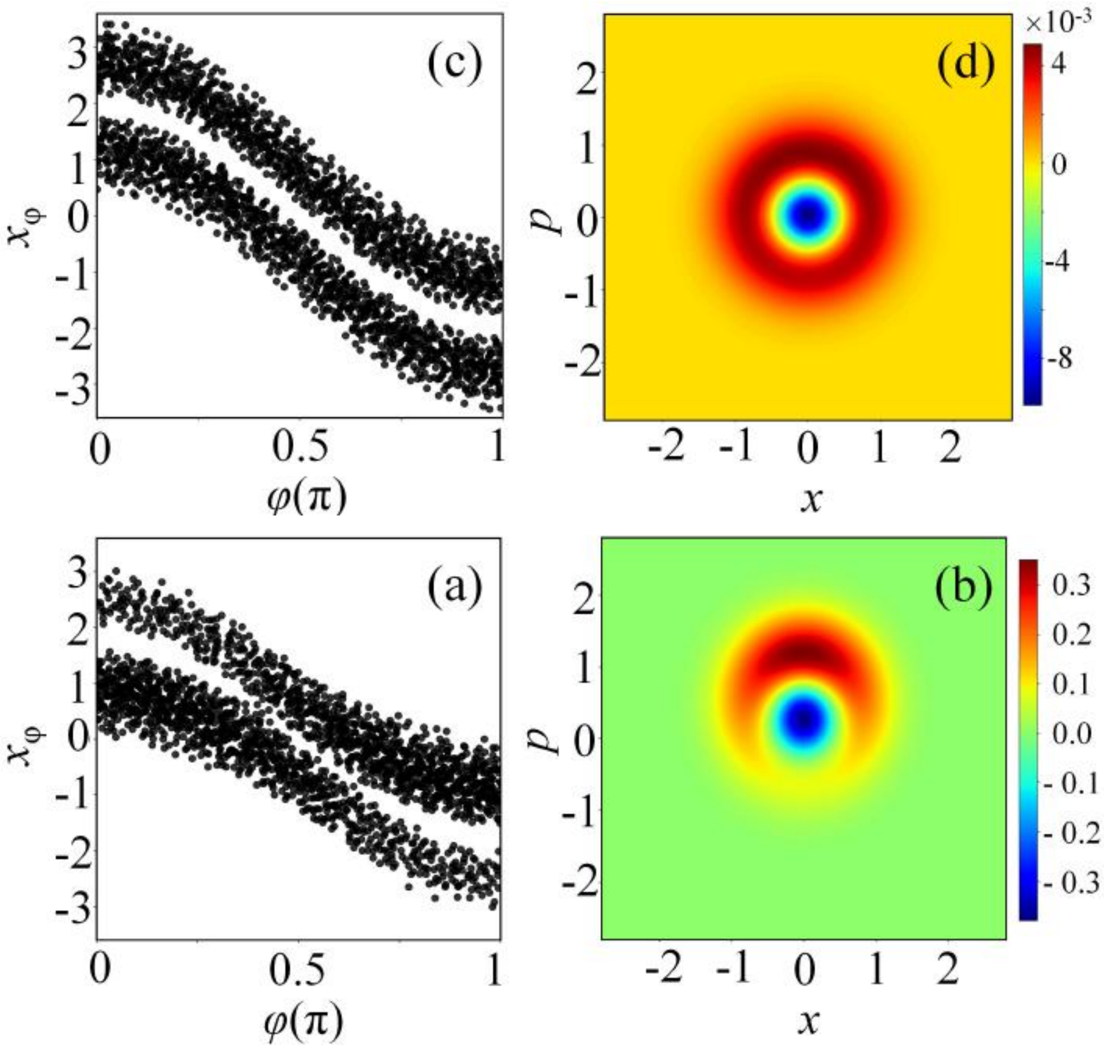


Gaussian distribution

Optical cat states: Wigner function

INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

Calculated Homodyne traces and reconstructed Wigner functions



Optical “kitten”

|cat>=|α_1>+ξ|α_2>

α_1=0.1, α_2=-0.2

Optical “cat”

|cat>=|α_1>+ξ|α_2>

α_1=0.3, α_2=-0.6

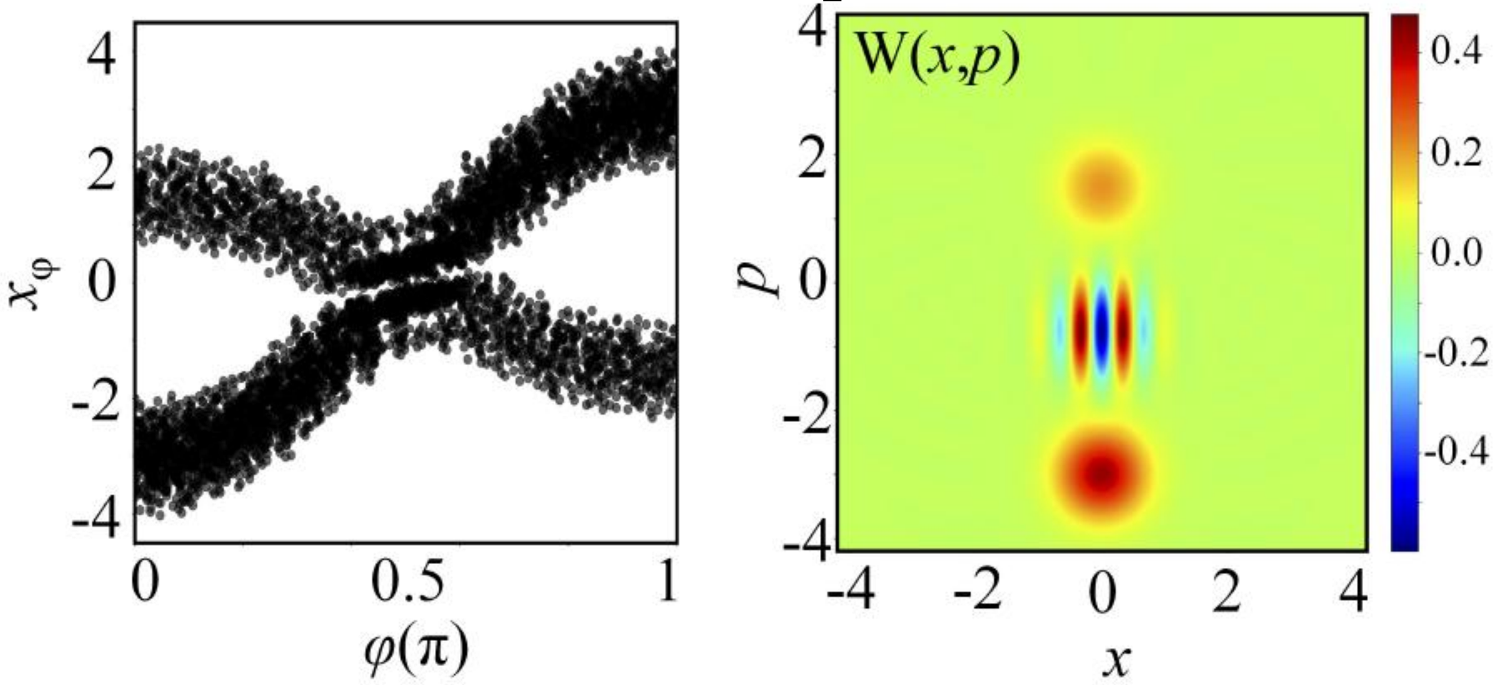
Optical cat states: Wigner function

"Large" optical cat states

$$|cat\rangle = |\alpha_1\rangle + \xi|\alpha_2\rangle$$

α_1=1.5, α_2=-3

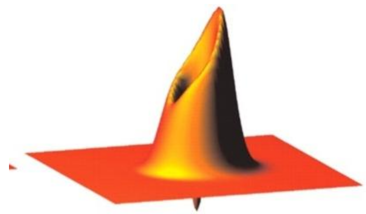
Calculated plots



Generation of optical cat states

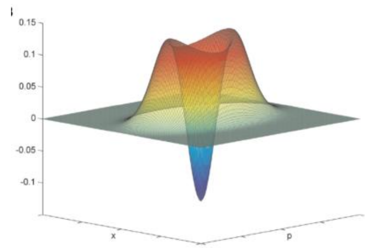
Enginiering optical "cat" states is not trivial

Single photon added coherent states



A. Zavata et al., *Science* **306**, 83, (2004)

Single-photon subtracted squeezed states



A. Ourjoumtsev et al., *Science* **312**, 83, (2006)

D. V. Sychev et al., *Nat. Photon.* (2017)

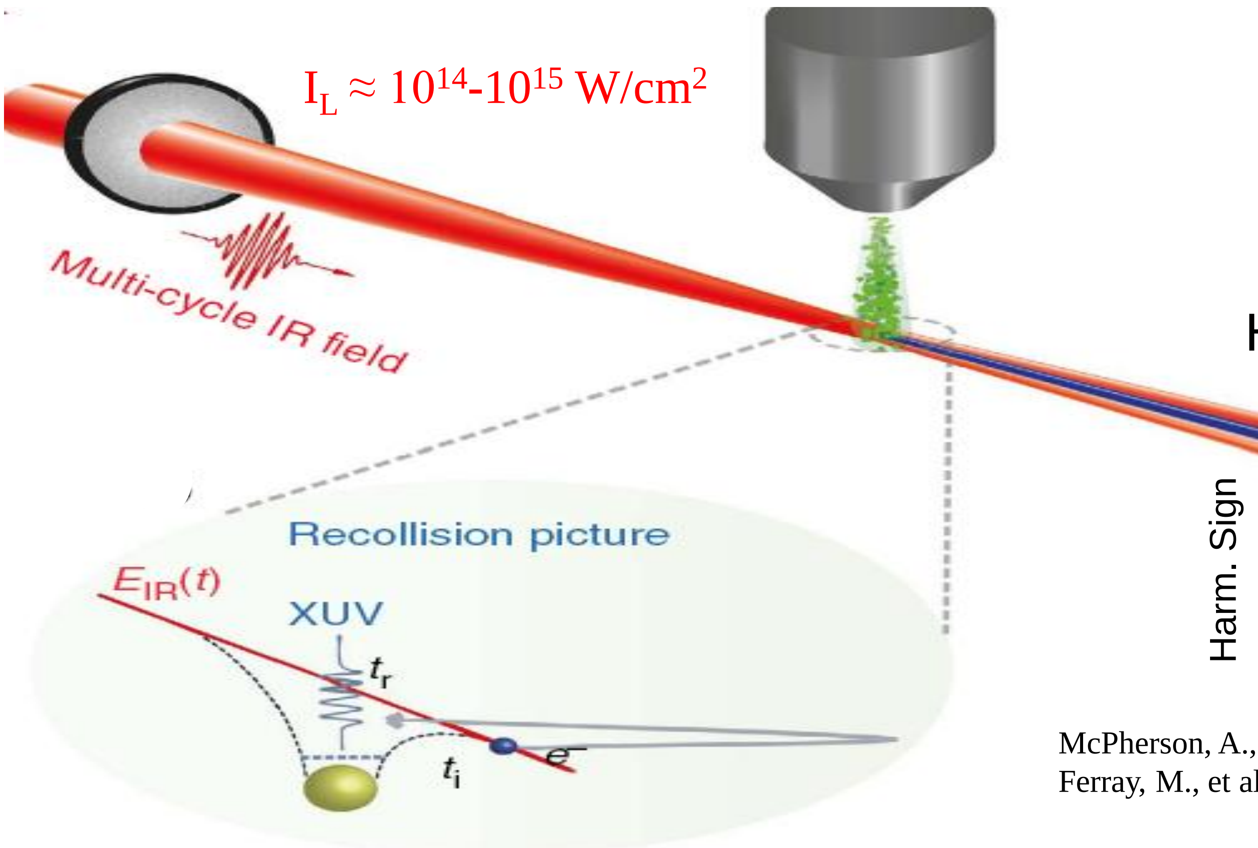
- Few photon number light sources

- The generation of high photon number optical cat states with controllable quantum features is considered of high importance

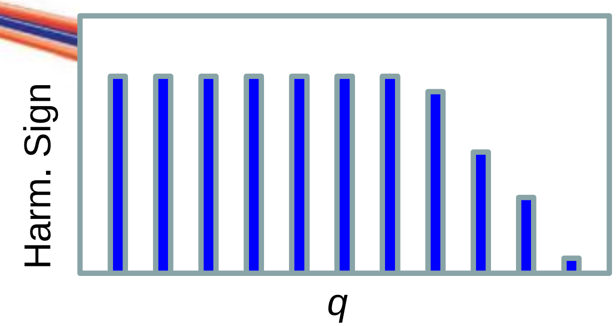
Generation of optical "cat" states using intense laser—atom interactions

FORTH
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

Strong field laser atom interaction and HHG: Semi-classical 3-step model



High harmonic spectrum



McPherson, A., et al., *J. Opt. Soc. Am. B* **4**, 595 (1987).
Ferray, M., et al., *J. Phys. B* **21**, L31 (1988).

K. J Schafer, *et al.* PRL. 70, 1599 (1993)

P. Corkum, PRL. 71, 1994 (1993)

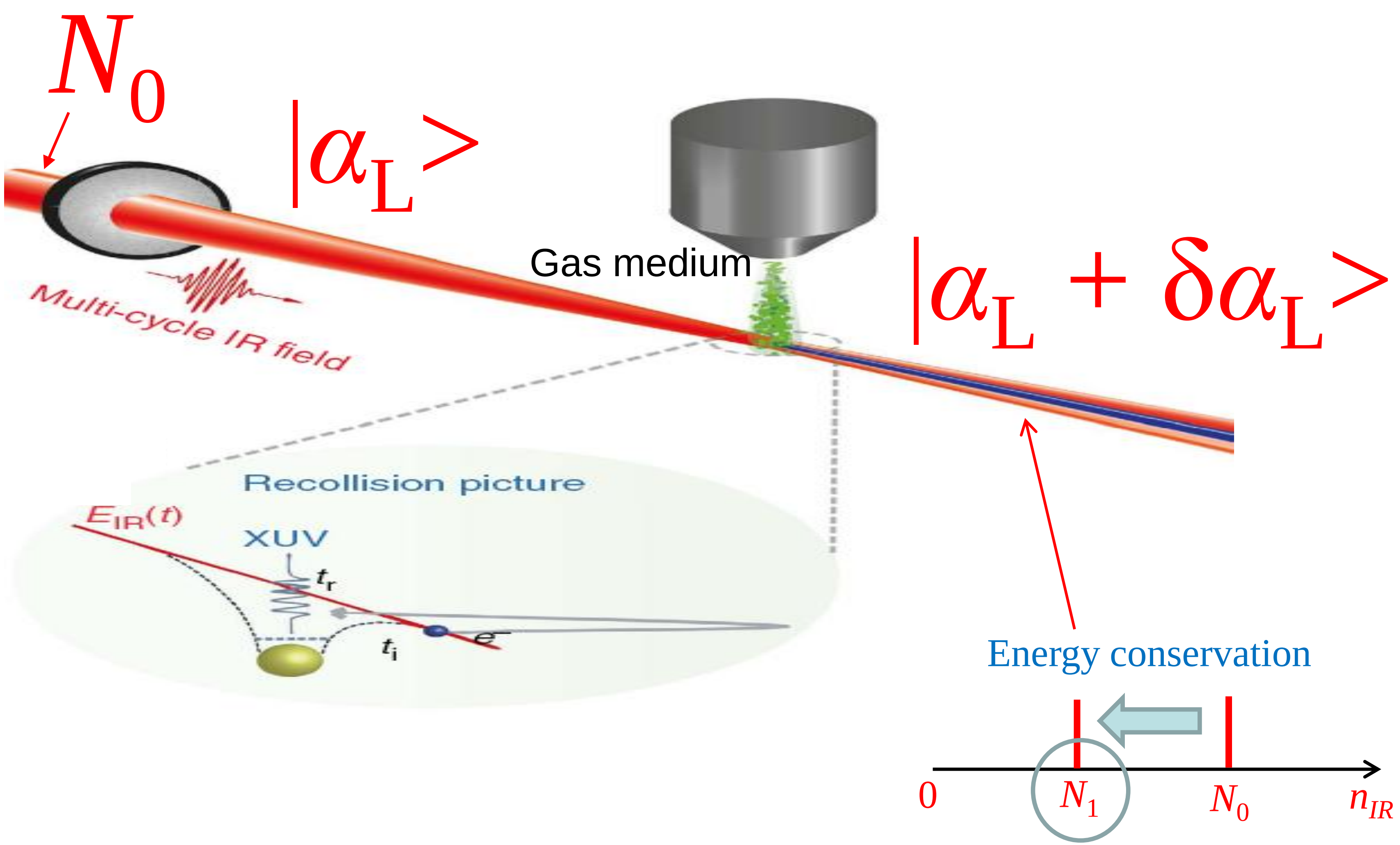
M. Lewenstein, *et al.* PRA. 49, 2117 (1994)

The EM radiation must be treated quantum mechanically i.e. the laser field using the coherent states of light

1) Which is the back-action of the interaction on the coherent state of the driving field ?

2) Which is the quantum state of the radiation after the interaction with atoms ?

Energy conservation

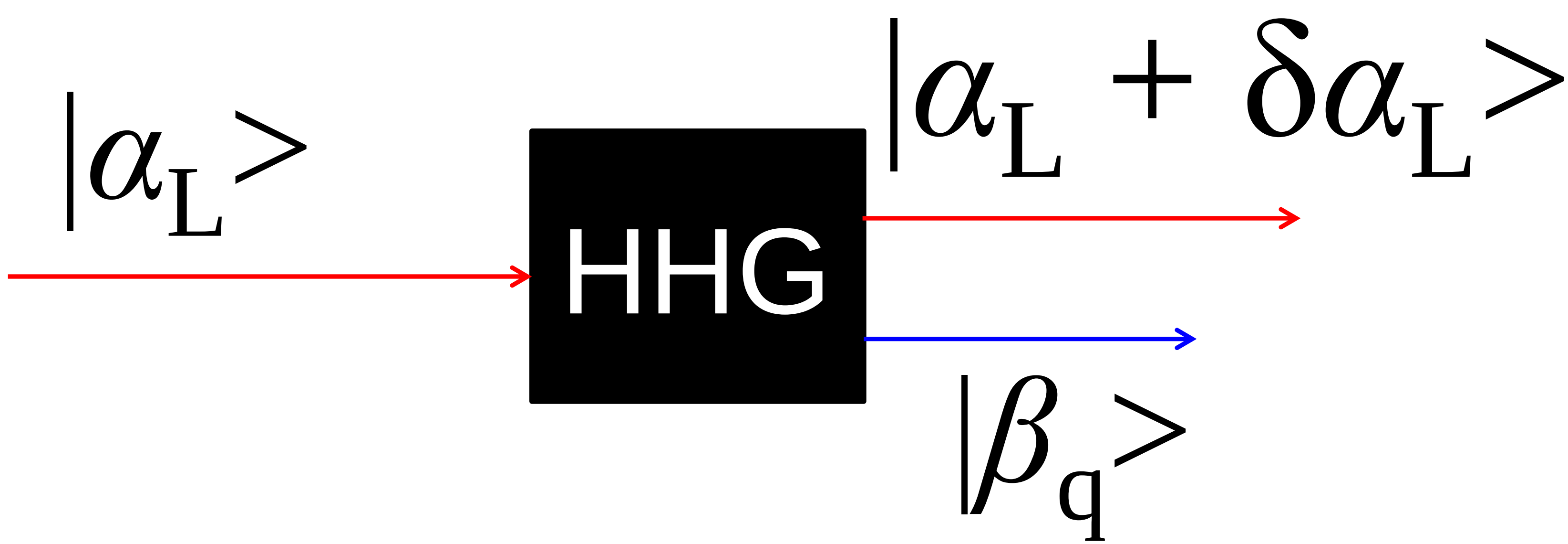


FORTH
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

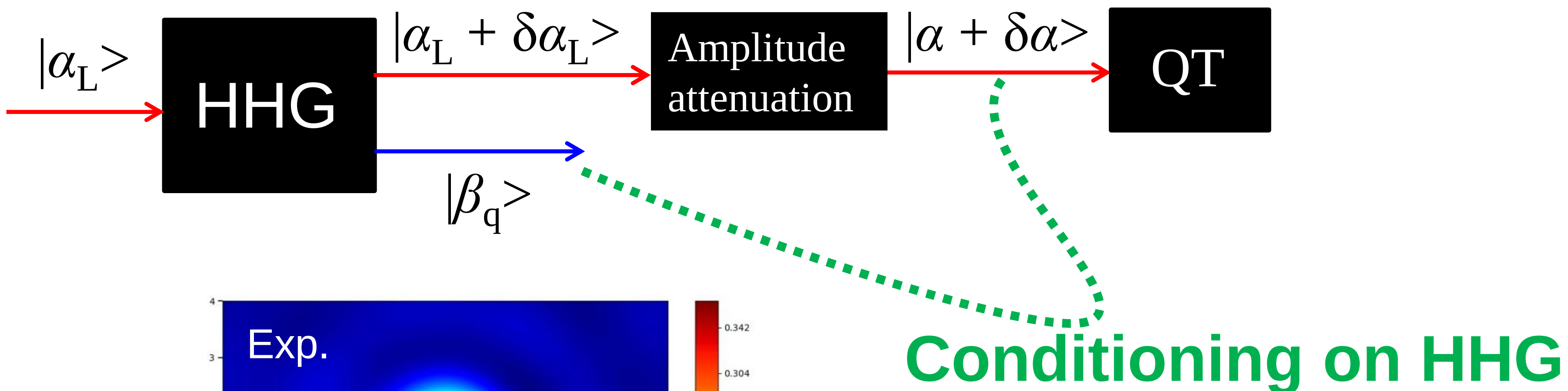
Back action of the HHG on the IR field

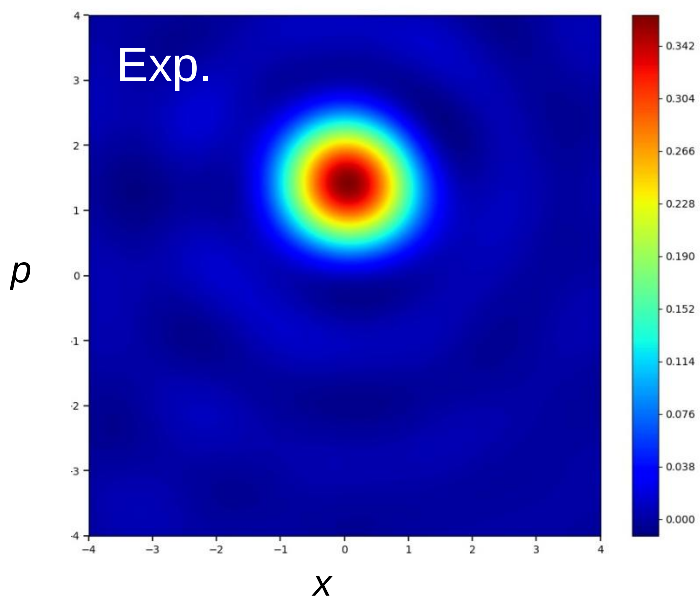
$$\text{TDSE: } i\hbar\frac{\partial}{\partial t}\left|\tilde{\Psi}(t)\right\rangle = \hat{H}\left|\tilde{\Psi}(t)\right\rangle \qquad \hat{H} = \hat{H}_0 + \hat{U} + \hat{H}_{\mathrm{f}}$$

$$\hat{U} = -e\hat{\mathbf{E}}\cdot\hat{\mathbf{r}} \qquad \hat{\mathbf{E}} = -i\hbar\mathbf{g}(\omega_{\mathrm{L}})f(t)\left[(\hat{a}^{\dagger} - \hat{a}) + \sum_{3}^{\mathrm{cutoff}}\sqrt{q}(\hat{b}_q^{\dagger} - \hat{b}_q)\right] \qquad \hat{H}_{\mathrm{f}} = \hbar\omega\hat{a}^{\dagger}\hat{a} + \sum_{q}^{\mathrm{cutoff}}\hbar\omega q\hat{b}_q^{\dagger}\hat{b}_q$$

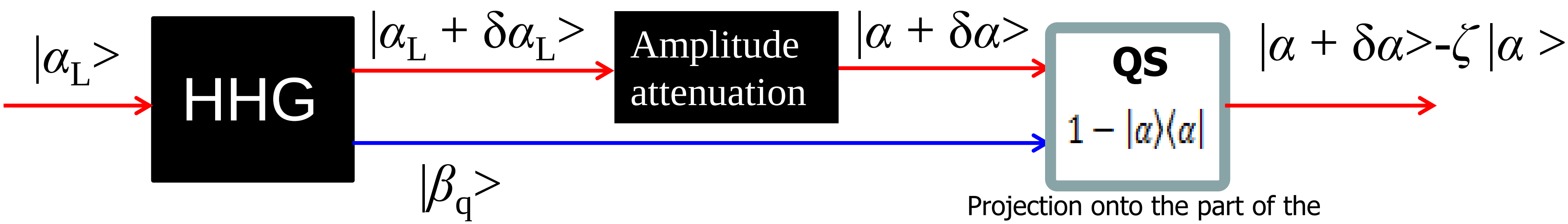


Back action of the HHG to the IR field

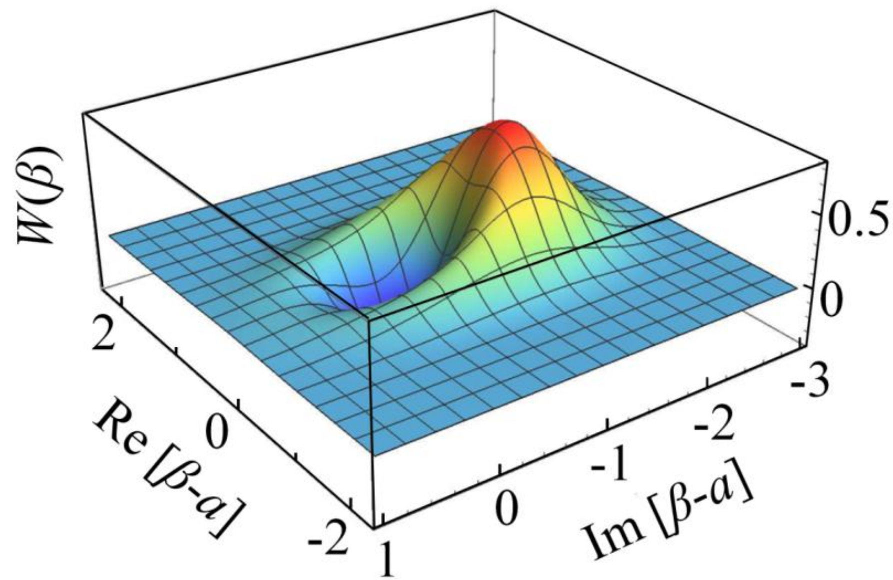


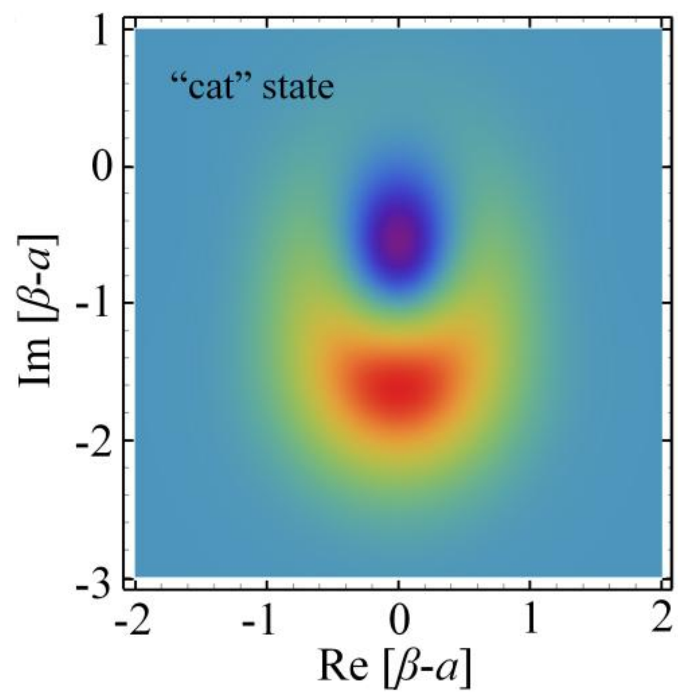


Conditioning on HHG



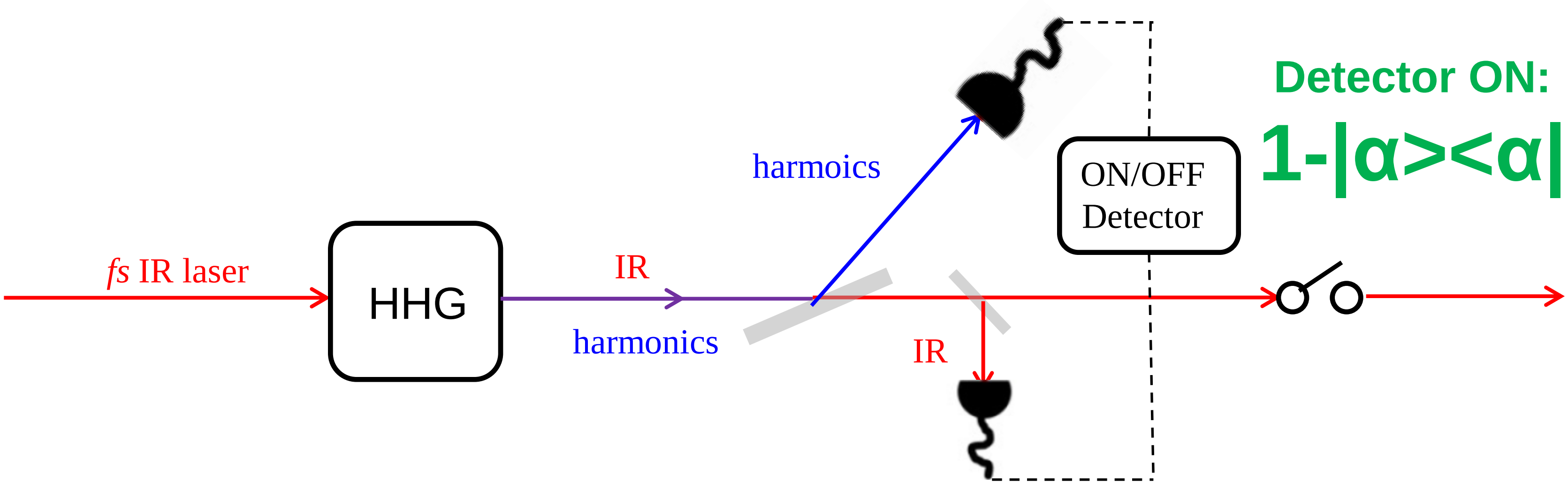
Projection onto the part of the fundamental that has been affected by the HHG process





M. Lewenstein *et. al.*, *Nature Phys.* **17**, 1104 (2021)

Conditioning on HHG



Detector ON:

1-|α><α|

Shot-to shot XUV/IR photon correlation method which selects (Detector ON) only the IR shots where the missing photons are relevant to the high harmonic generation.

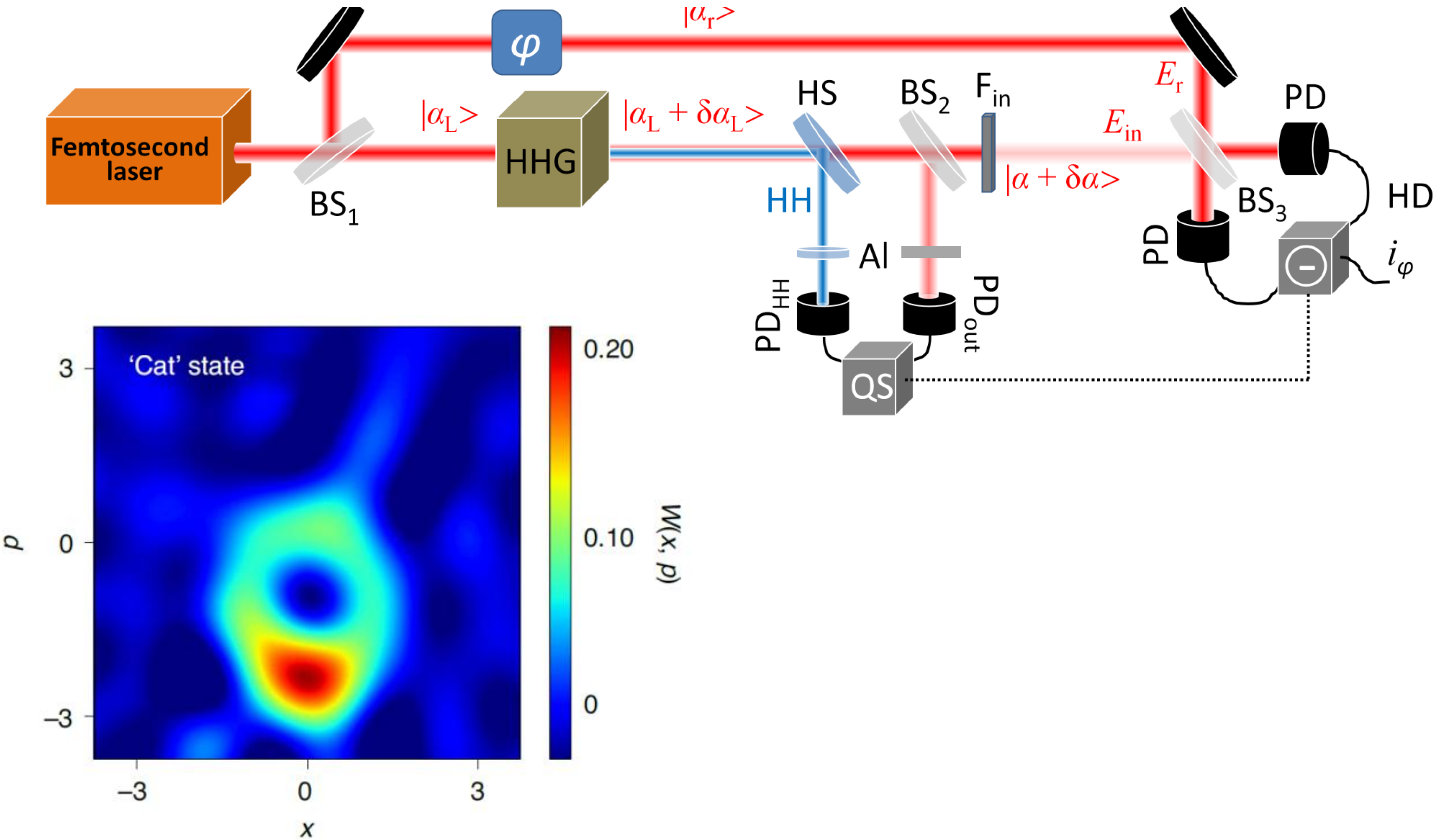
N. Tsatrafyllis, et al., *Nature Commun.* 8, 15170 (2017)

M. Lewenstein *et. al.*, *Nature Phys.* 17, 1104 (2021)

J. Rivera-Dean *et. al.*, PRA 105, 033714 (2022)

P. Stammer et al., *PRX Quantum,* **4**, 010201 (2023)

Quantum state characterization



M. Lewenstein *et. al.*, *Nature Phys.* **17**, 1104 (2021)

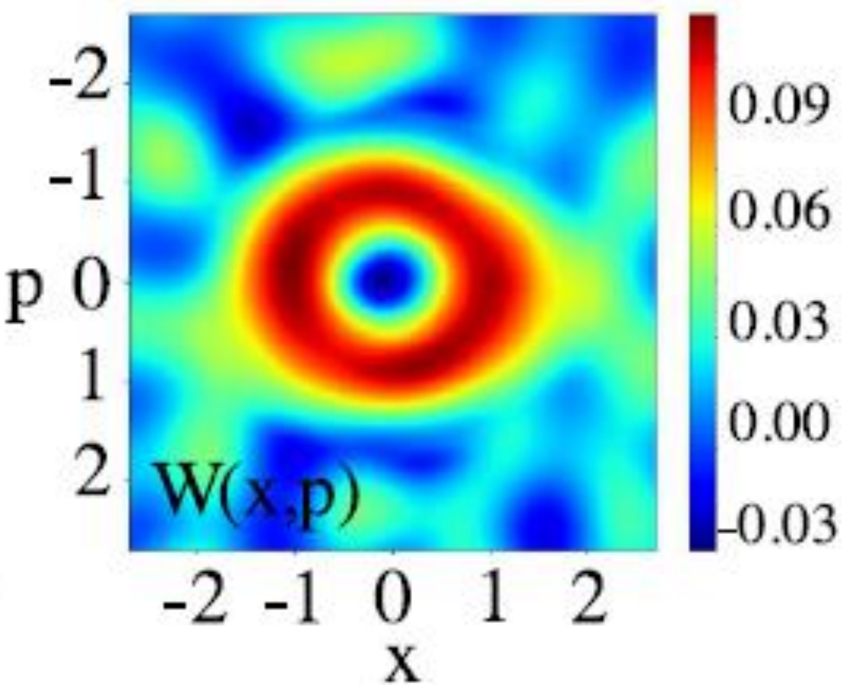
Generation of high photon number shifted optical cat states with controllable quantum features

Control of the cat states

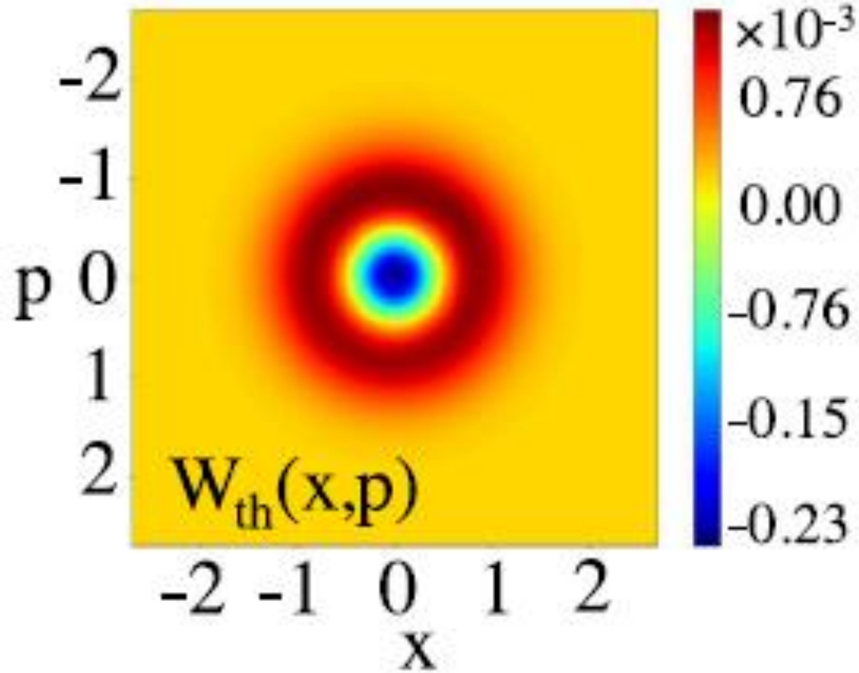
"Controlling" the quantum features of the "cat" states

$|Cat\rangle = |\alpha + \delta\alpha\rangle - \zeta|\alpha\rangle$ **for $\delta\alpha \ll \alpha$ (low N)**

Experiment



Theory

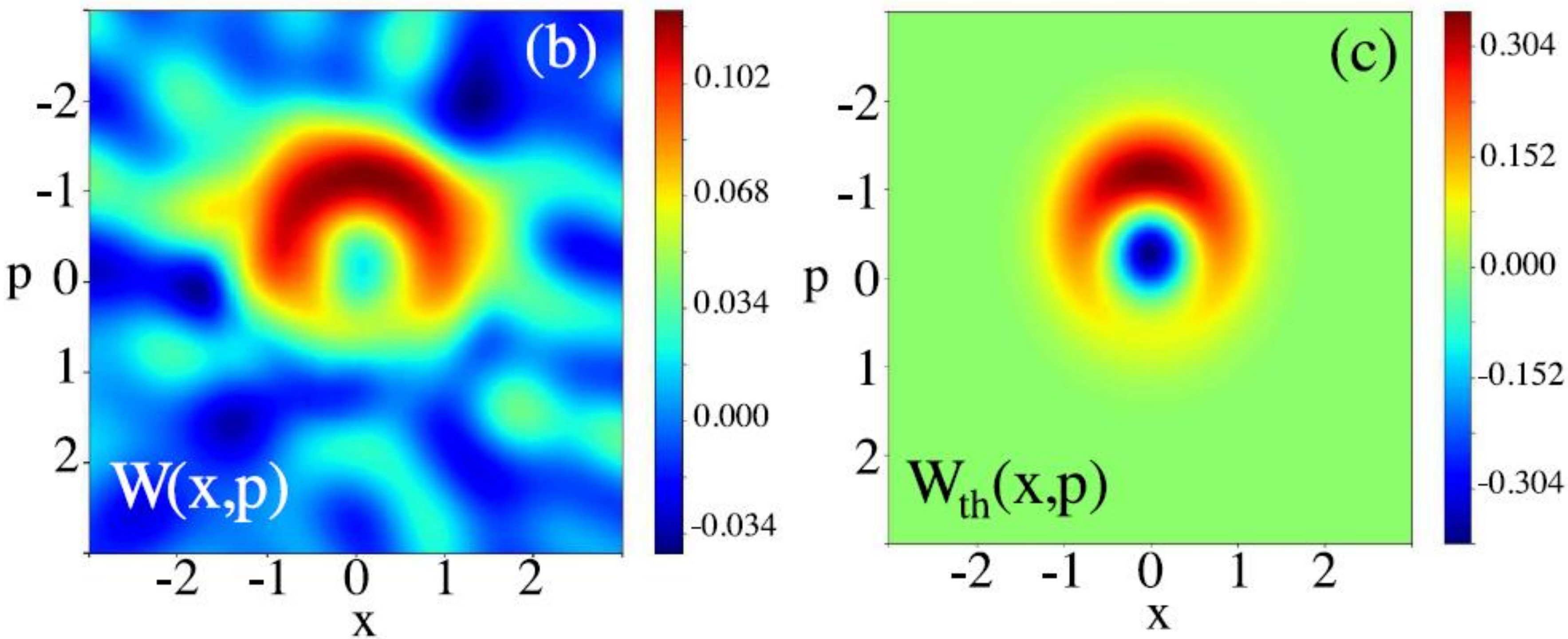


J. Rivera-Dean *et. al.*, PRA **105,** 033714 (2022)

High photon number cat states

Generation of high photon number optical “cat” states

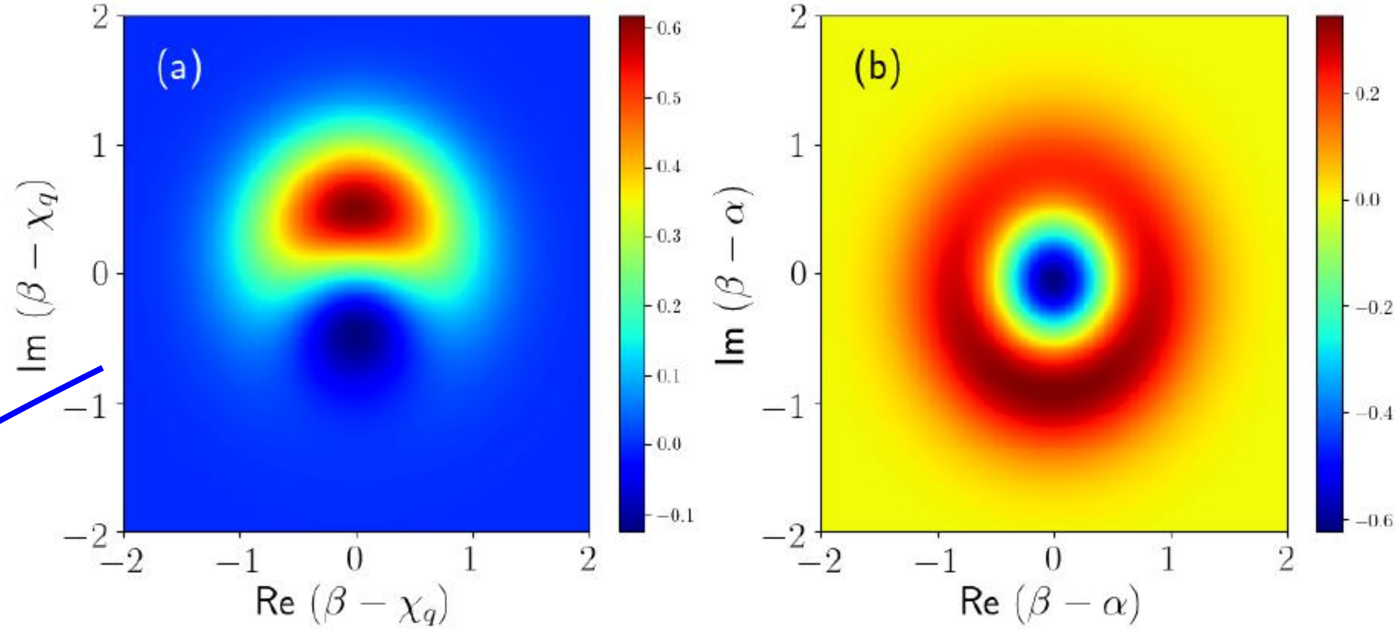
9-photon shifted optical “cat” states



J. Rivera-Dean *et. al.*, PRA **105,** 033714 (2022)

Theoretical results

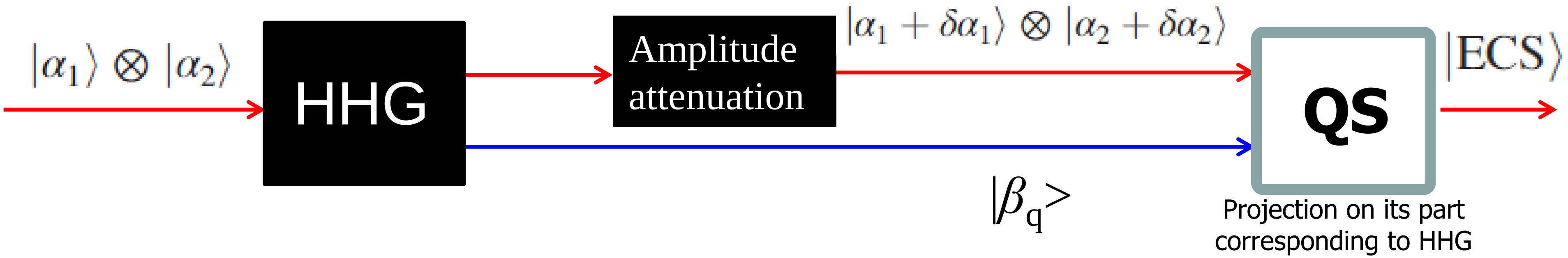
HH IR



Experimentally can be done using the harmonics generated in Vis-IR spectral range

P. Stammer et al., *PRL* **128**, 123603 (2022)

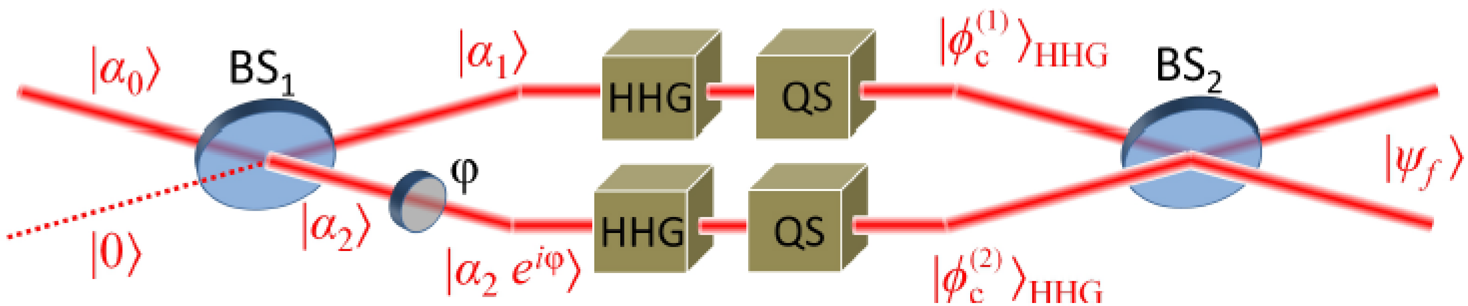
Two-color driving field



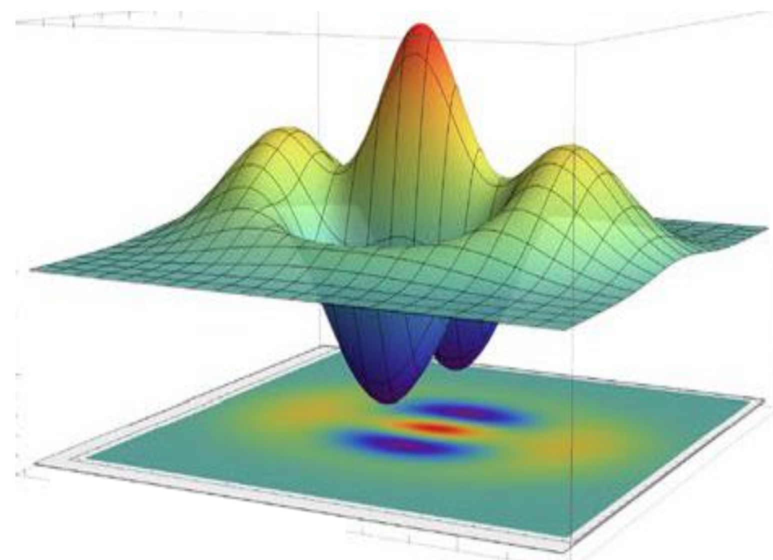
$$|ECS>=|\alpha_1+\delta\alpha_1>|\alpha_2+\delta\alpha_2>+\xi\,|\alpha_1>|\alpha_2>$$

P. Stammer et al., *PRL* **128**, 123603 (2022)

New schemes for creating large optical cat and entangled states



$$
\begin{aligned}
|\psi_f\rangle = & \left| \frac{1}{\sqrt{2}}[\alpha_1(1+e^{i\varphi}) + \chi_1 + \chi_2 e^{i\varphi}] \right\rangle_t \\
& \otimes \left| \frac{1}{\sqrt{2}}[-\alpha_1(1-e^{i\varphi}) + \chi_1 + \chi_2 e^{i\varphi}] \right\rangle_r \\
& + \xi_2 \left| \frac{1}{\sqrt{2}}[\alpha_1(1+e^{i\varphi}) + \chi_1] \right\rangle_t \\
& \otimes \left| \frac{1}{\sqrt{2}}[-\alpha_1(1-e^{i\varphi}) + \chi_1] \right\rangle_r \\
& + \xi_1 \left| \frac{1}{\sqrt{2}}[\alpha_1(1+e^{i\varphi}) + \chi_2 e^{i\varphi}] \right\rangle_t \\
& \otimes \left| \frac{1}{\sqrt{2}}[-\alpha_1(1-e^{i\varphi}) + \chi_2 e^{i\varphi}] \right\rangle_r \\
& + \xi_1 \xi_2 \left| \frac{1}{\sqrt{2}}[\alpha_1(1+e^{i\varphi})] \right\rangle_t \\
& \otimes \left| \frac{1}{\sqrt{2}}[-\alpha_1(1-e^{i\varphi})] \right\rangle_r .
\end{aligned}
$$



J. Revera-Dean et al., *J. Comput. Electr.* **20**, 2111 (2021)

P. Stammer et al., *PRX Quantum*, **4**, 010201 (2023)

Non-linear optics using intense optical "cat" states

Nonliner optics using intense optical Schrödinger "cat" states

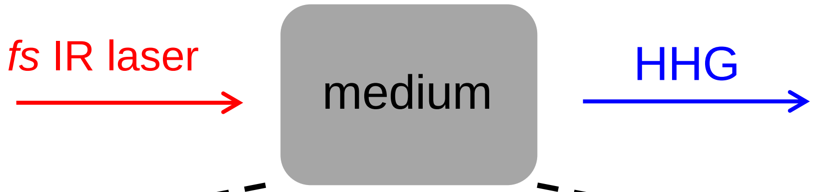
Th. Lamprou,[1,2†], J. Rivera-Dean[3†], P. Stammer[3†], M. Lewenstein[3,4], and P. Tzallas[1,5*]

arXiv:2306.14480v1 [quant-ph] 26 Jun 2023

FORTH
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

Conclusions

The findings are just the beginning of a very long "story"



Molecules, crystals, Nanostructures, 2-D materials, laser-plasma interactions

↓

New class of non-classical light sources

↓

Applications to quantum technology

Conclusions

Strong laser physics, non–classical light states and quantum information science

U. Bhattacharya[1], Th. Lamprou[2,3], A. S. Maxwell[4], A. Ordóñez[1], E. Pisanty[5], J. Rivera-Dean[1], P. Stammer[1], M. F. Ciappina[6,7,8], M. Lewenstein[1,9] and P. Tzallas[2,10*]

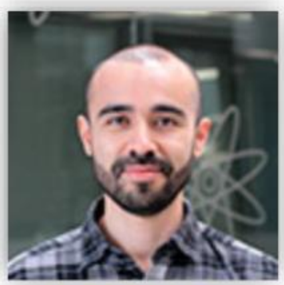
arXiv:2302.04692v1 [quant-ph] 9 Feb 2023

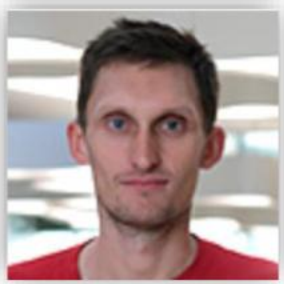
Acknowledgments:

ICFO


M. Lewenstein


J. Rivera-Dean


A. F. Ordóñez


P. Stammer

FORTH
INSTITUTE OF ELECTRONIC STRUCTURE AND LASER


P. Tzallas


T. Lamprou


E. Skantzakis

AARHUS UNIVERSITY


A. S. Maxwell

KING'S College LONDON


E. Pisanty


M. F. Ciappina


THANK YOU