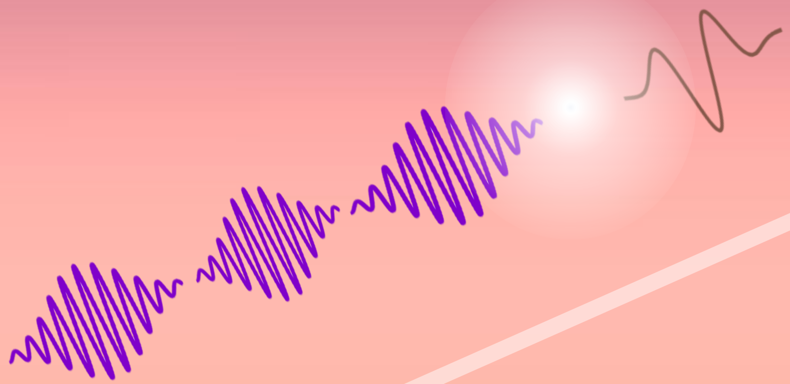
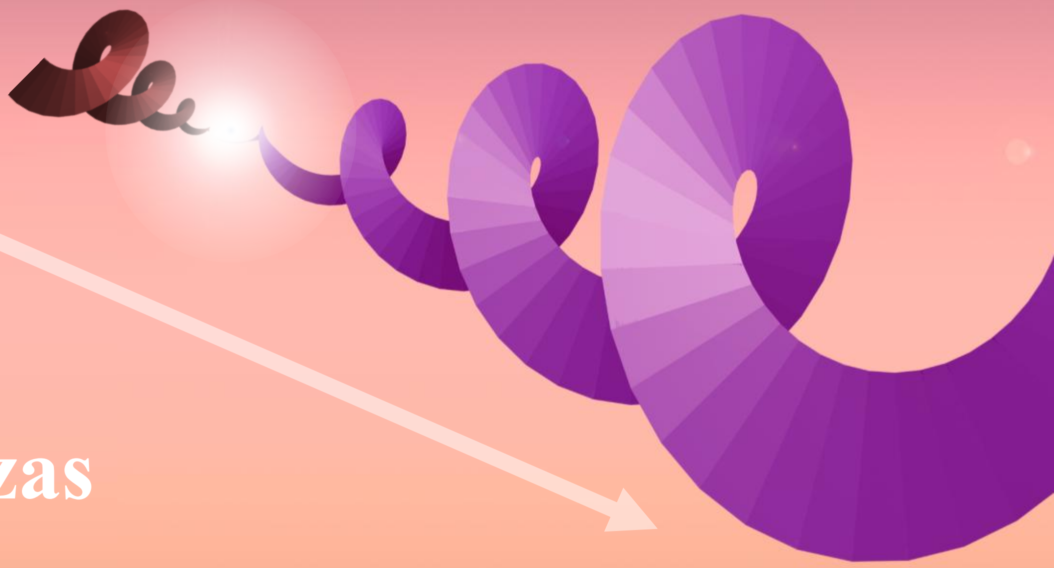
# Structuring high-order harmonic generation with angular momentum: from fundamentals to applications





Laura Rego Cabezas

IUPAP prize talk, ICAP 2026, Wuhan

CSIC icmm

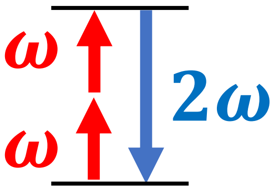
# Fundamentals of high-order harmonic generation

# High-order harmonic generation

- The investigation of ultrafast mechanisms in Nature benefits from the use of laser pulses with two characteristics: **ultrashort duration and high frequency.**

- Such light pulses can be created via **non-linear optics**.

M. Göppert-Mayer. Annalen der Physik 401, 273–294 (1931)
W. Kaiser, et al. Physical Review Letters 7, 229–231 (1961)
P. A. Franken, et al. Physical Review Letters 7, 118–119 (1961)




$$P = \epsilon_0\left(\chi^{(1)}E(\omega) + \chi^{(2)}E(\omega)^2 + \chi^{(3)}E^3 + \cdots\right)$$

Second harmonic generation

# High-order harmonic generation

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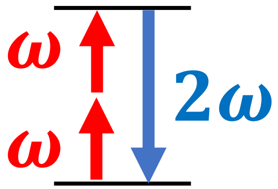


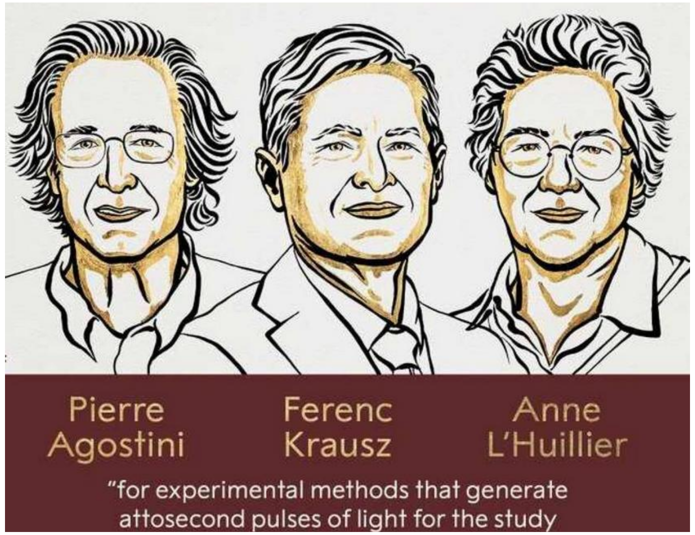

$$P = \epsilon_0(\chi^{(1)}E(\omega) + \chi^{(2)}E(\omega)^2 + \chi^{(3)}E^3 + \cdots)$$

Second harmonic generation

- The development of intense lasers reached enough intensity to observe **high-order harmonic generation (HHG).**

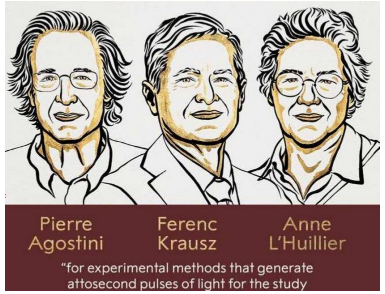
A. McPherson, et al. Journal of the Optical Society of America B 4, 595 (1987)
M. Ferray, et al. Journal of Physics B: Atomic, Molecular and Optical Physics 21, L31–L35 (1988)
P. M. Paul, et al. Science 292, 1689–1692 (2001)
M. Hentschel, et al. Nautre 414, 6863, 509-514 (2001)

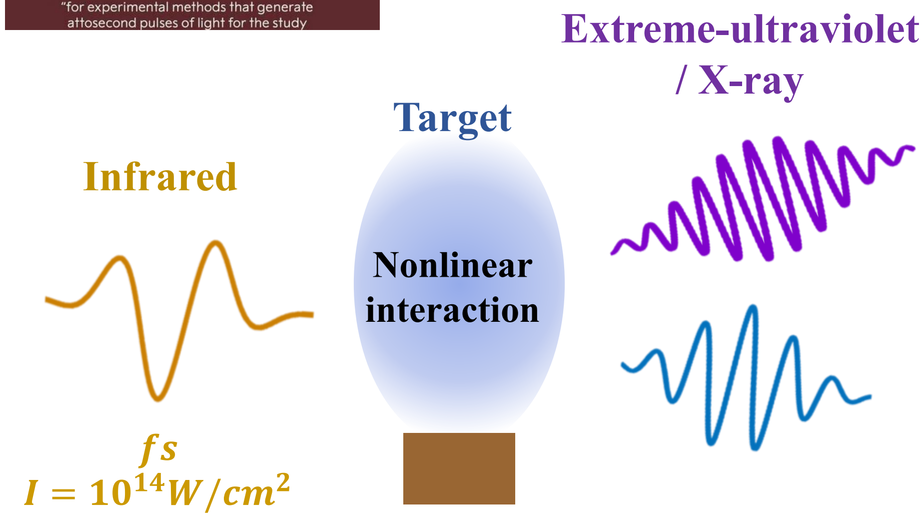
Nobel Prize in Physics 2023



# High-order harmonic generation

- High-order harmonic generation is a highly nonlinear and **nonperturbative** frequency conversion phenomenon.



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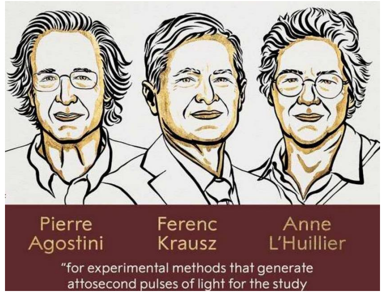
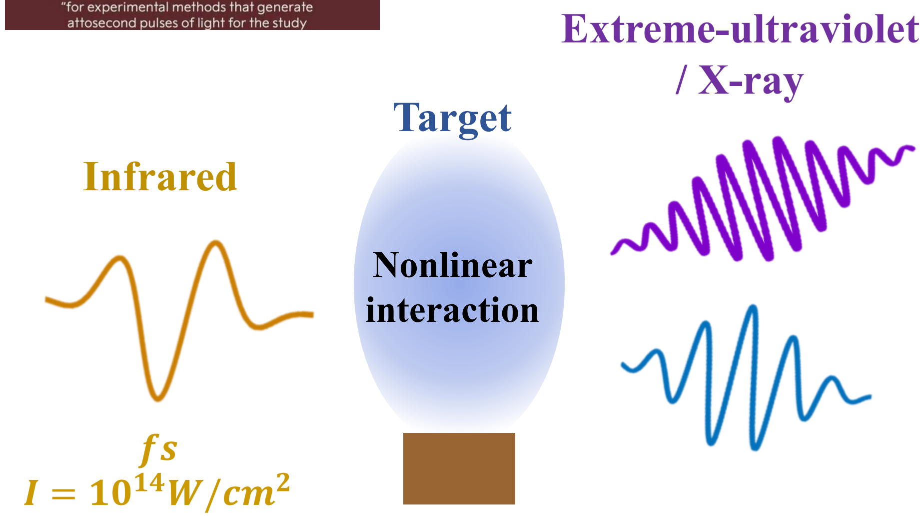
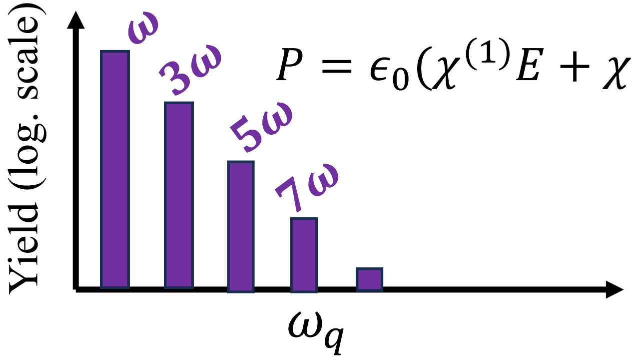



- Perturbative




$$P = \epsilon_0(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \cdots)$$

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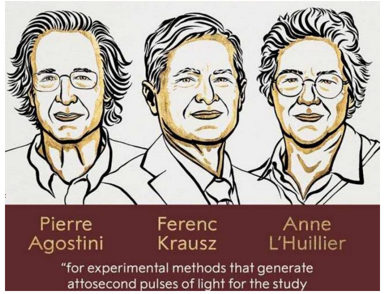


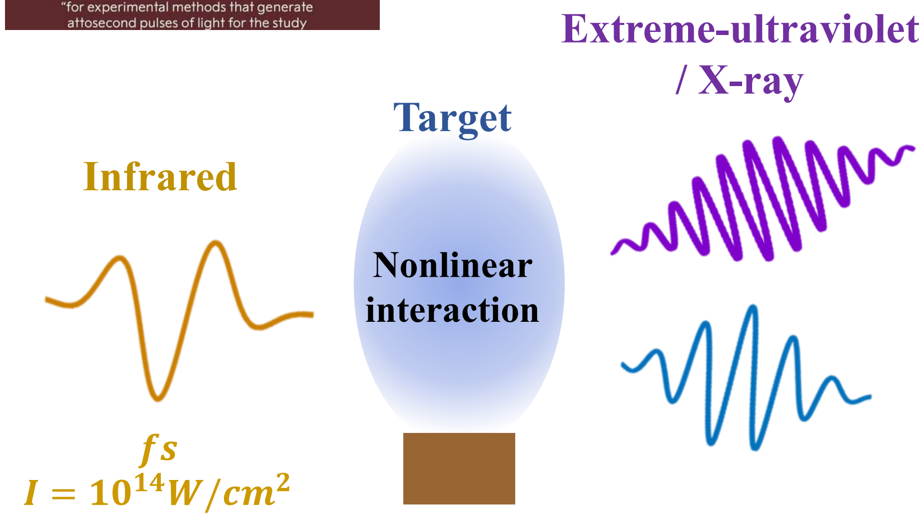


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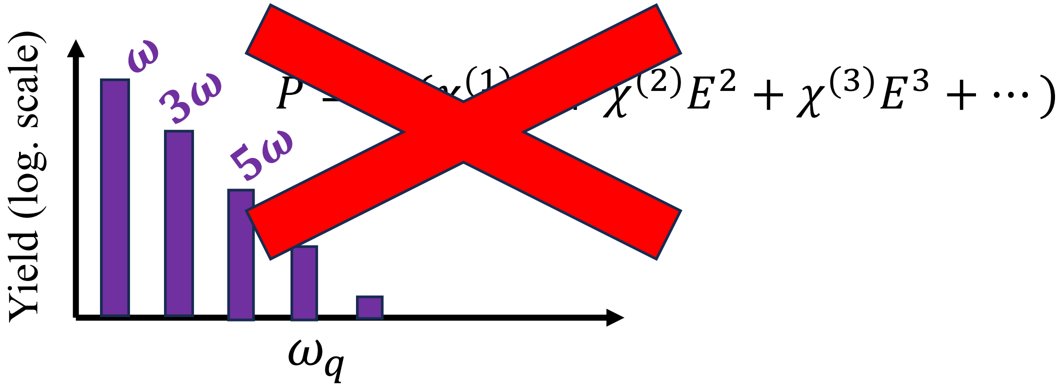
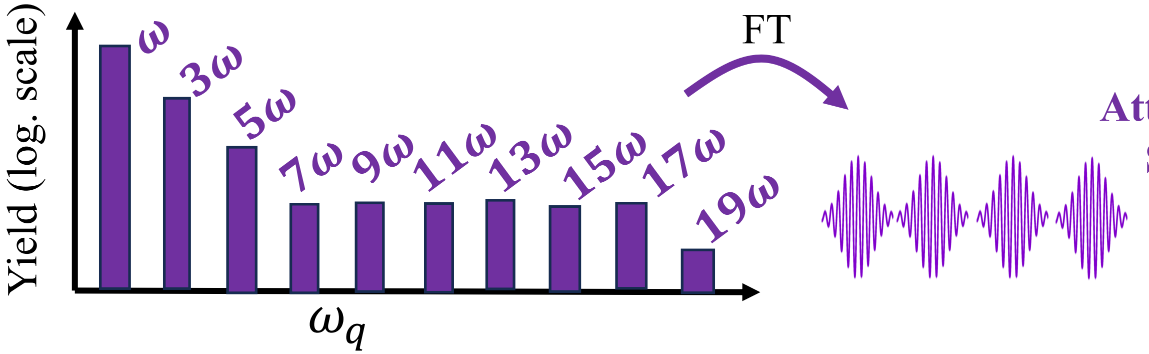



- Nonperturbative




**Attosecond pulses! Shortest laser pulses**

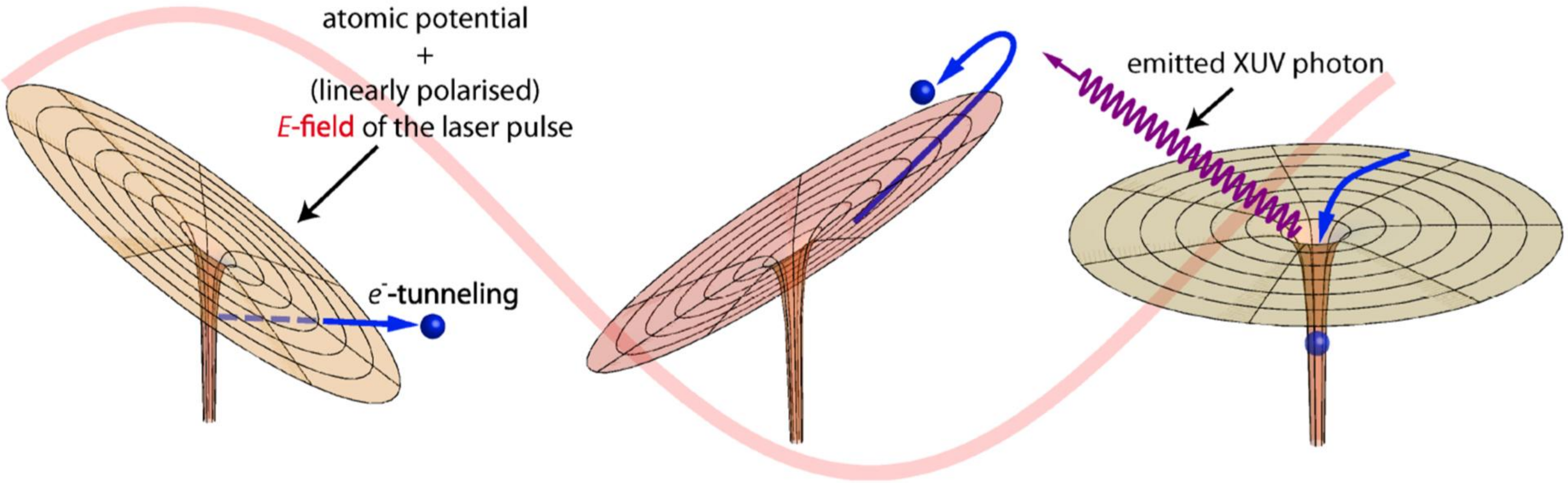
$1\ as = 10^{-18} s$

# High-order harmonic generation

- Microscopically, HHG can be described using a simple 3 step model.

K. J. Schafer, et al. Physical Review Letters 70, 1599–1602 (1993)
P. B. Corkum, Physical Review Letters 71, 1994–1997 (1993)

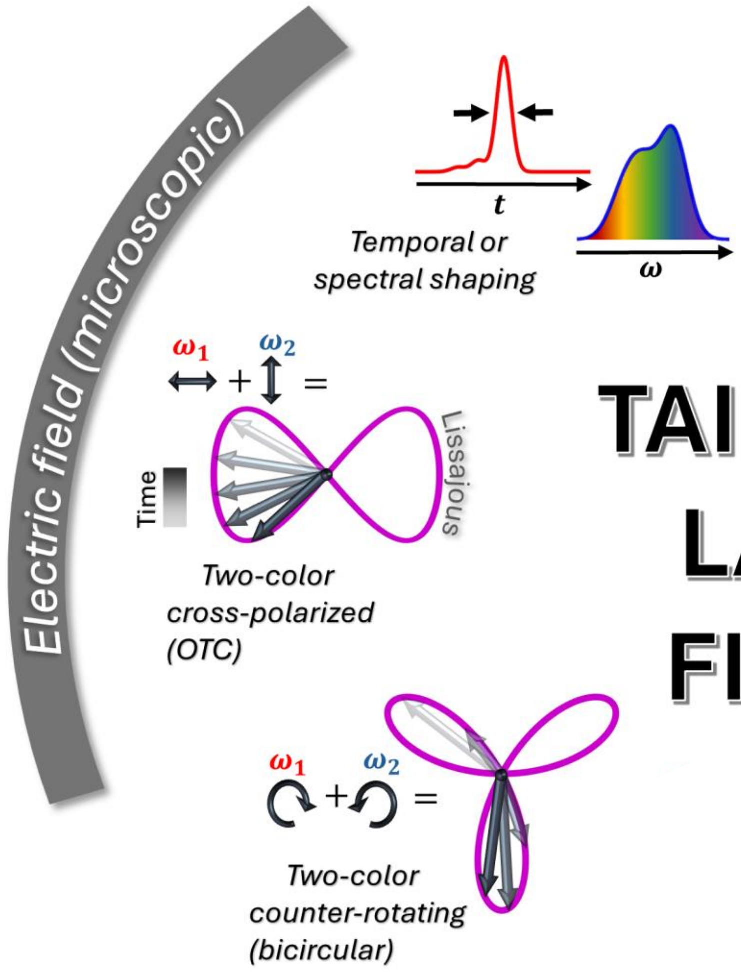
1. Tunnel ionization
2. Acceleration
3. Recombination




© C. Kern, et al. Nanophotonics 4, 303–323 (2015)

# Structured light and the angular momentum of light

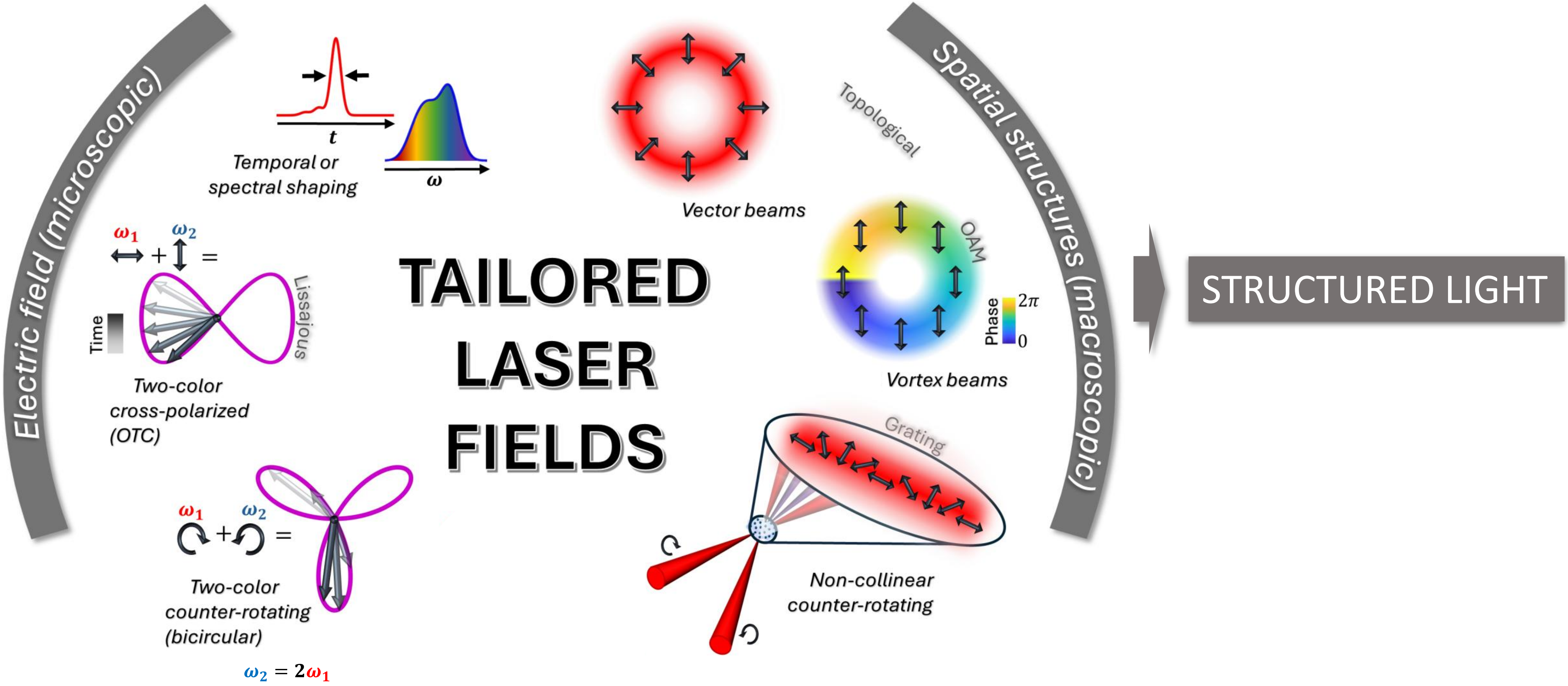
# How can you tailor light?




$\omega_2 = 2\omega_1$

**TAILORED LASER FIELDS**

# How can you tailor light?



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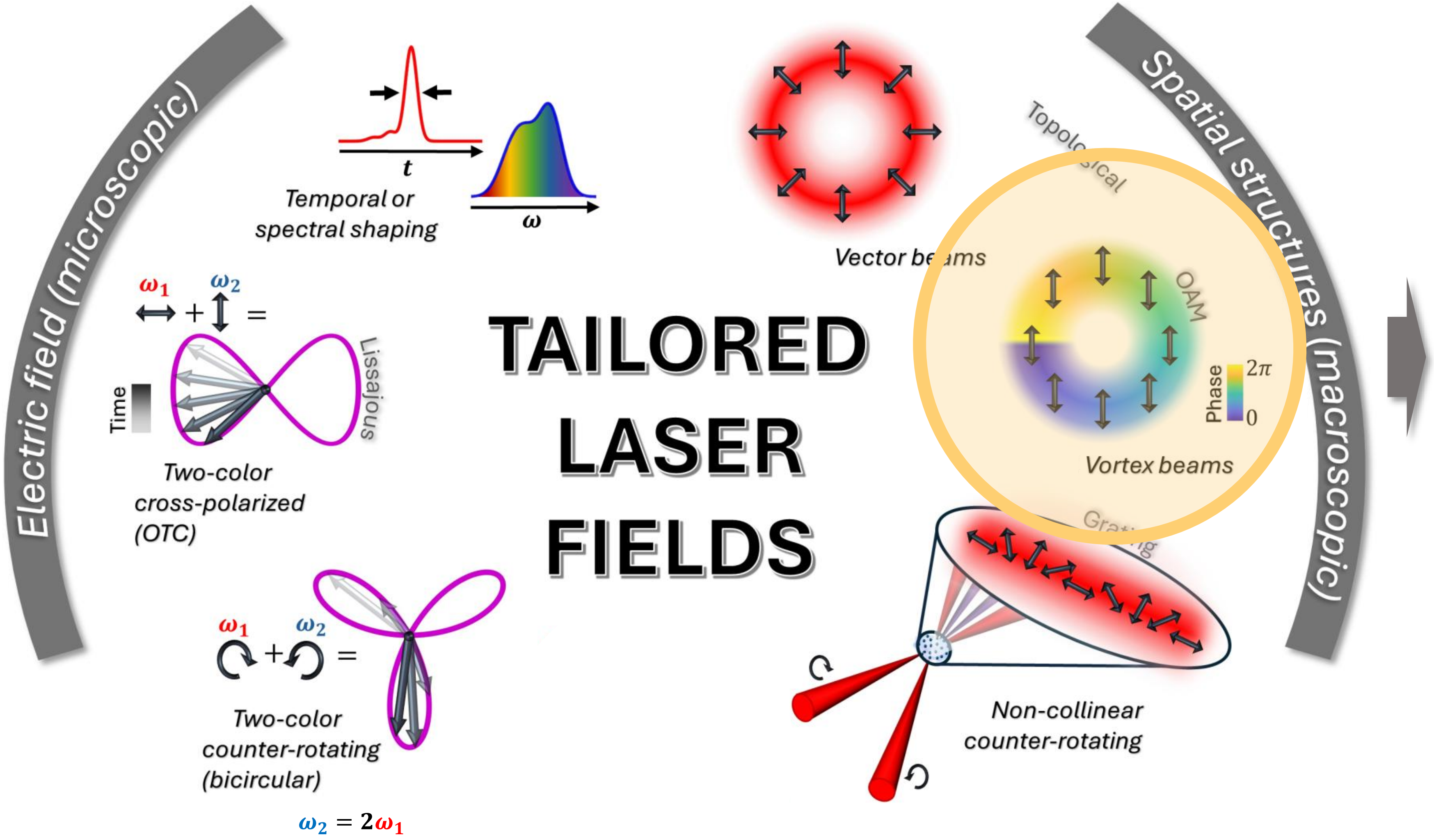
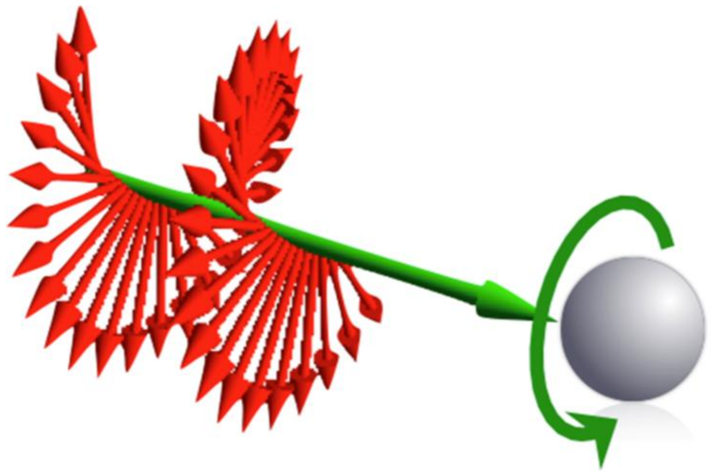



STRUCTURED LIGHT

# The angular momentum of light

- Spin Angular Momentum (SAM)

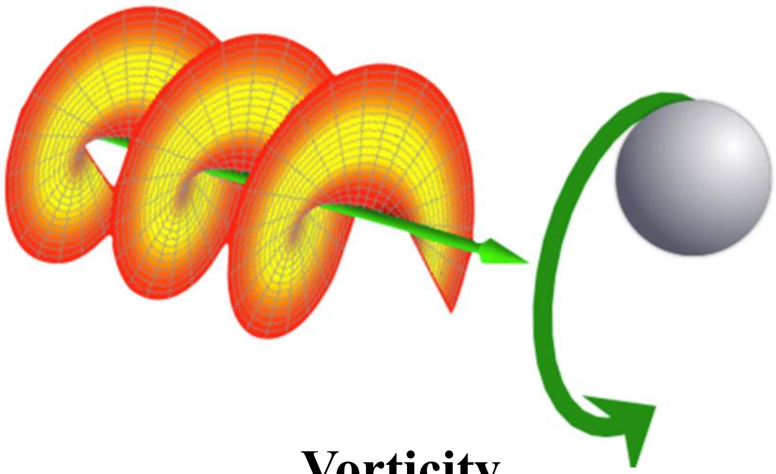


**Polarization**

Microscopic

$\sigma = \pm 1$

- Orbital Angular Momentum (OAM)



**Vorticity**

Macroscopic

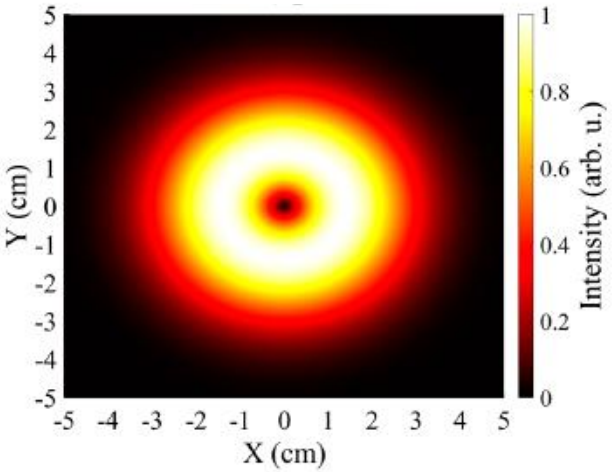
$\ell = 0, \pm 1, \pm 2, \pm 3, \pm 4 \ldots$

# The orbital angular momentum of light

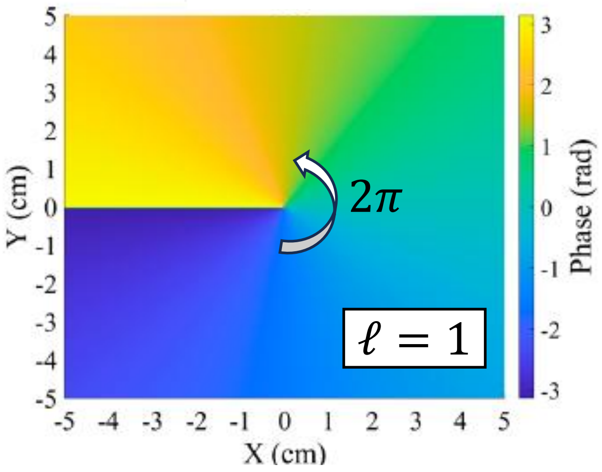
- ❑ Vortex beams carry orbital angular momentum

**L. Allen, et al. Physical Review A, 45, 8185 (1992)**

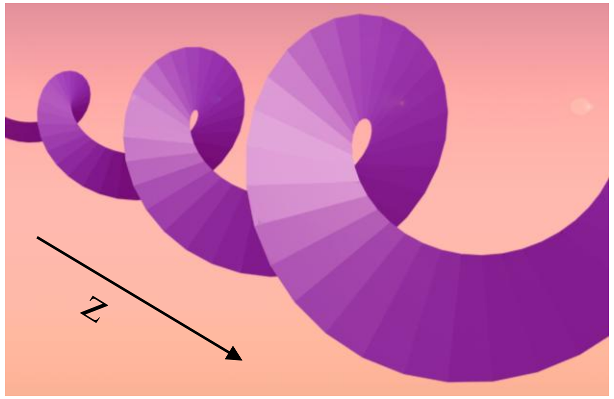
Intensity




Phase




Wavefront



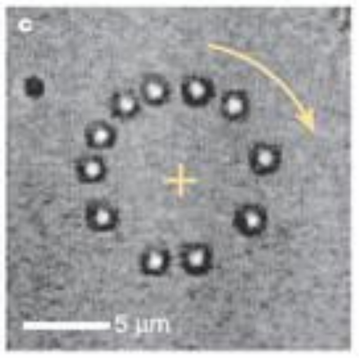

$$\ell = \frac{1}{2\pi} \times \oint_C \nabla\phi(\mathbf{r})\mathrm{dr}$$

OAM: number of $2\pi$ shifts of the phase along the azimuth

# The orbital angular momentum of light

- Vortex beams carry orbital angular momentum
- They are mathematically described by Laguerre Gauss modes
- They exhibit very promising applications:

- A vortex beam can transfer its orbital angular momentum to matter, imprinting a rotation into micromachines




*A micrometric particle trapped in a vortex beam rotates!*

**D.G. Grier, et al. Nature 424, 21 (2006)**

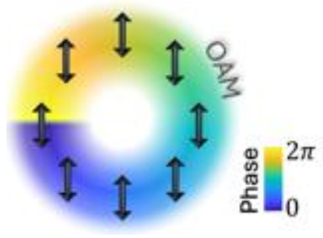
- Orbital angular momentum is a new degree of freedom that can be used to transfer information

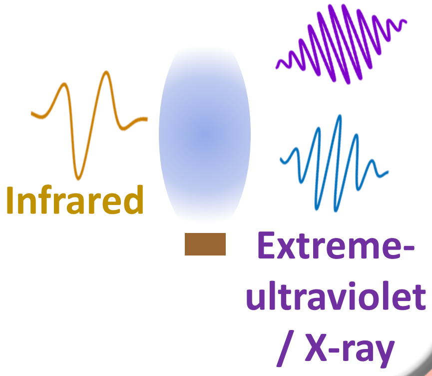
**J. Wang, et al. Nature Photonics 6, 488 (2012)**

- Additionaly, vortex beams have applications in fundamental physics and microscopy

**A. Mair, et al. Nature 412, 313–316 (2001)**

Structured light + High-order harmonic generation




## Control over emitted light

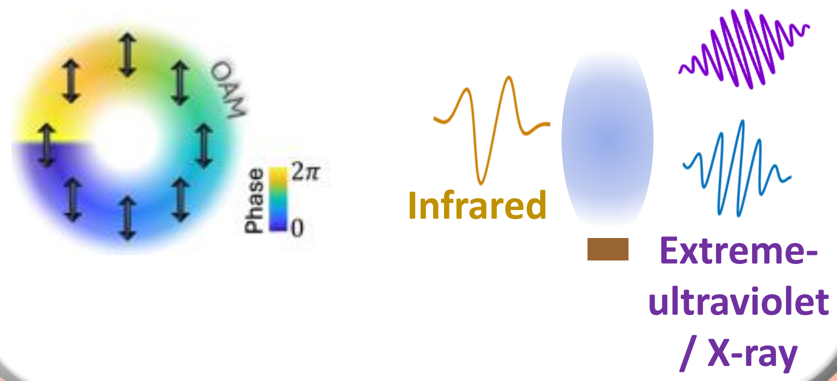
= 
- ❑ Extreme-ultraviolet / X-ray structured light
- ❑ New types of structured light: self-torque of light

=
- ❑ Control over other properties of the extreme-ultraviolet/X-ray light: polarization or spectral content.

## Spectroscopy

=
- ❑ Information about the target: chirality

**Structured light** + **High-order harmonic generation**

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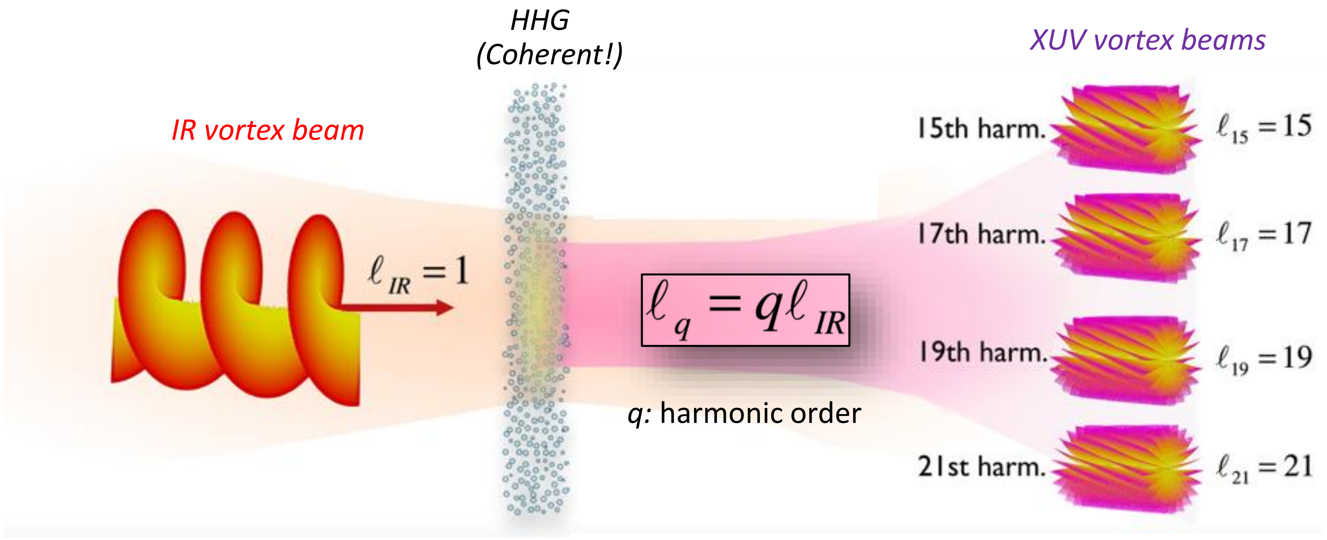
# The generation of extreme-ultraviolet vortex beams

- Extreme-ultraviolet (XUV) vortex beams are emitted from HHG driven by an infrared (IR) vortex beam





M. Zürch, et al. Nature Physics 8, 743 (2012)
C. Hernández García, et al. Physical Review Letters 111, 083602 (2013)
G. Gariepy, et al. Physical Review Letters 113, 153901 (2014)
R. Géneaux, et al. Nature Communications 7, 12583 (2016)

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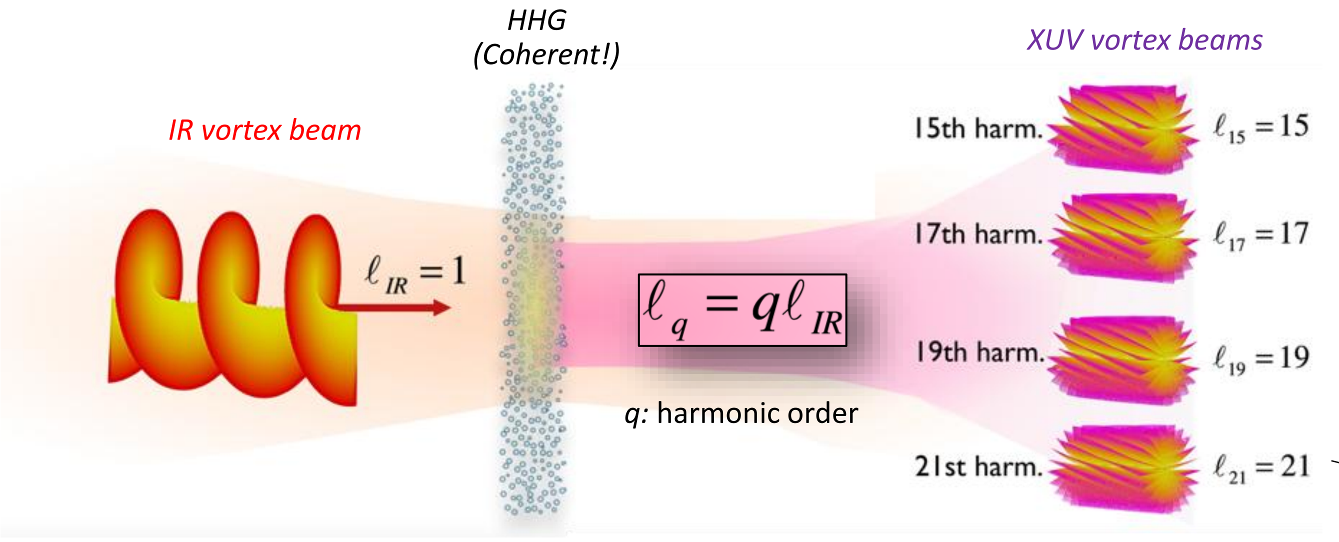
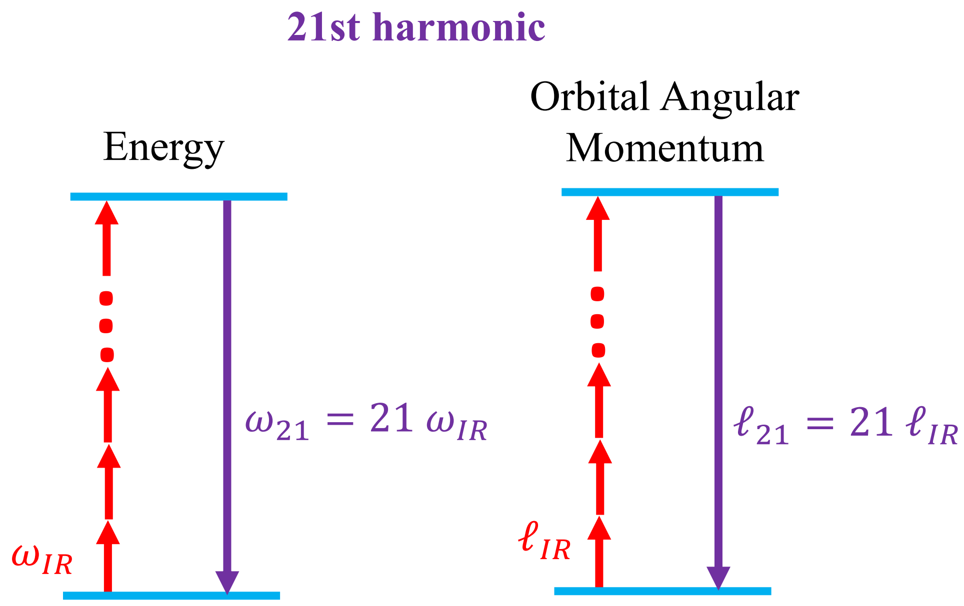



The orbital angular momentum of the harmonics follows a simple conservation rule:

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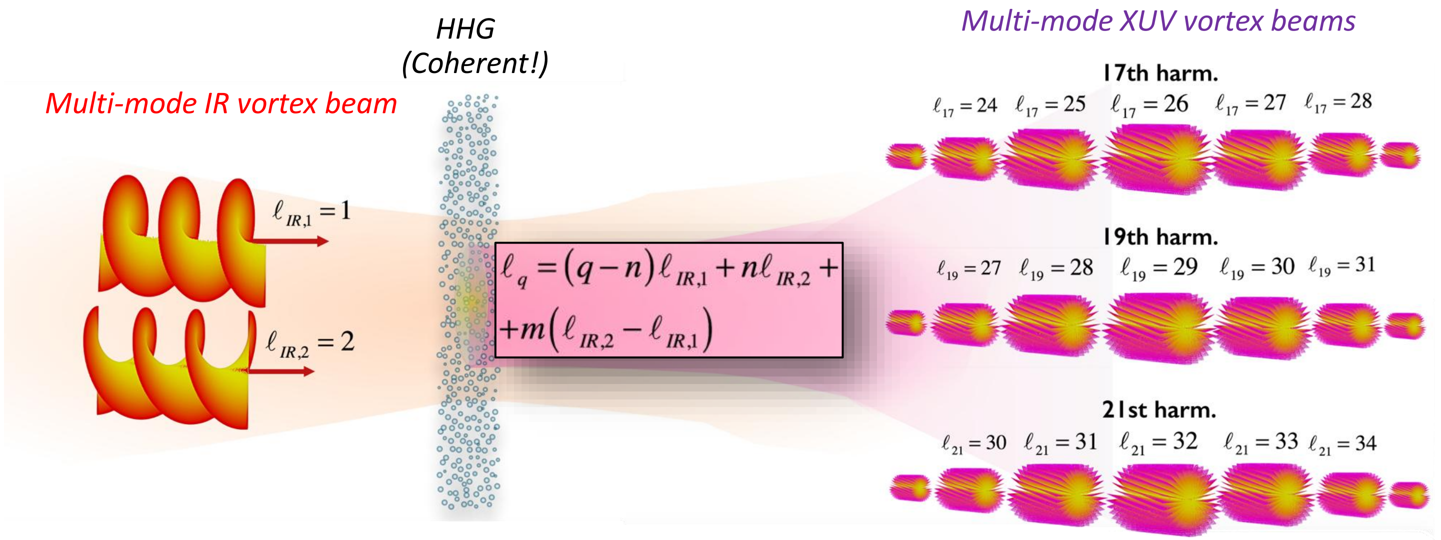
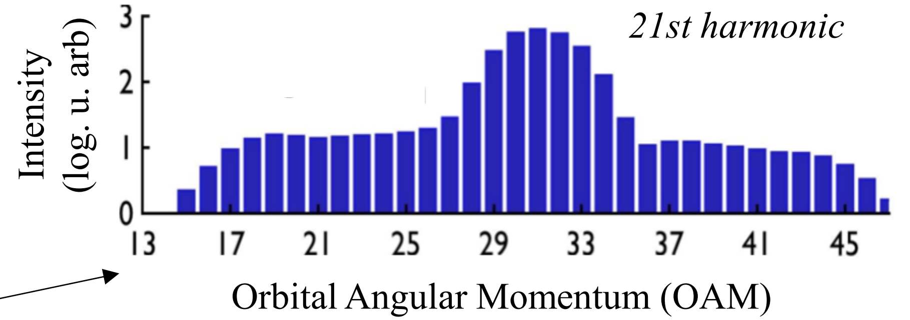
# Combination of vortex beams

- When combining several infrared vortex beams the resulting harmonics exhibit a broad orbital angular momentum content




Orbital angular momentum spectrum





VNiVERSiDAD D SALAMANCA
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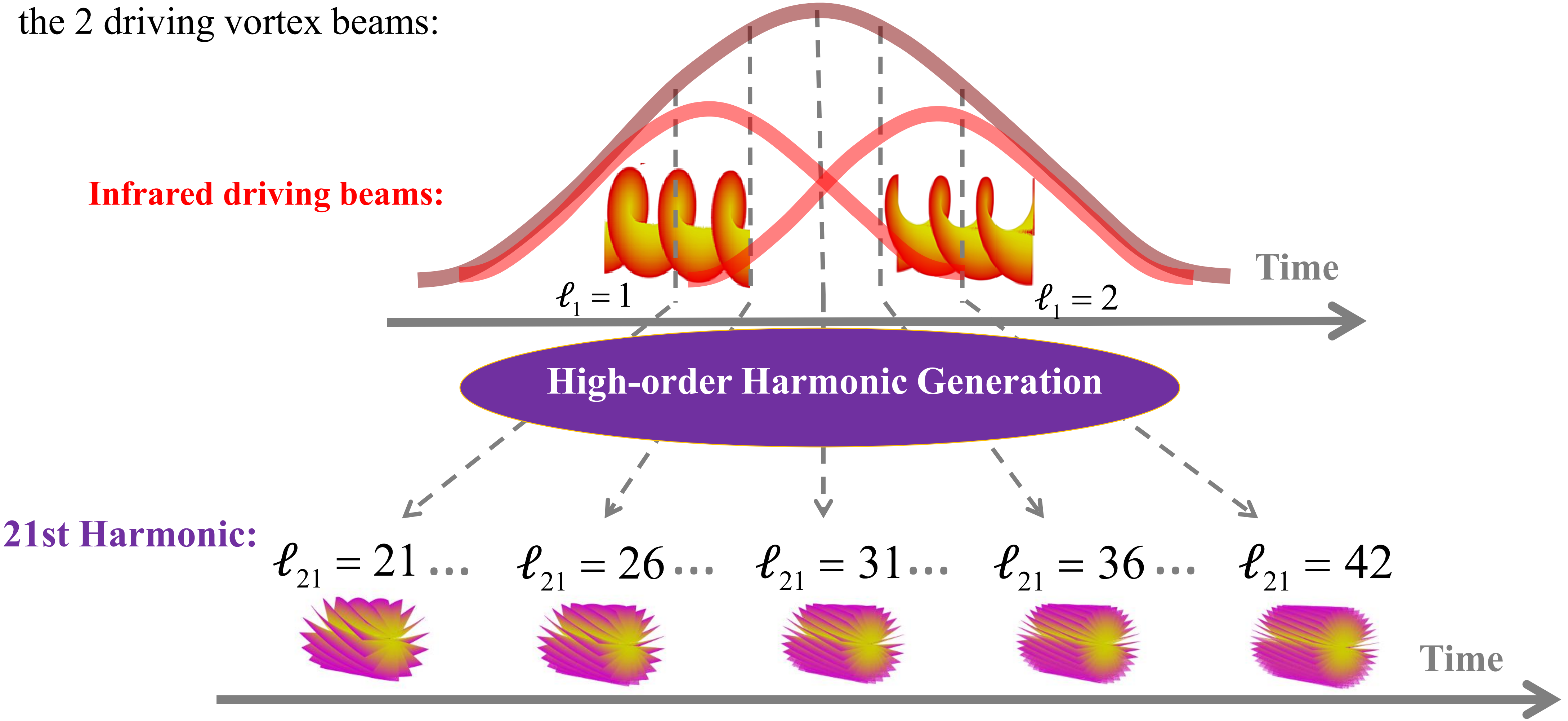
Argonne NATIONAL LABORATORY
A. Picón

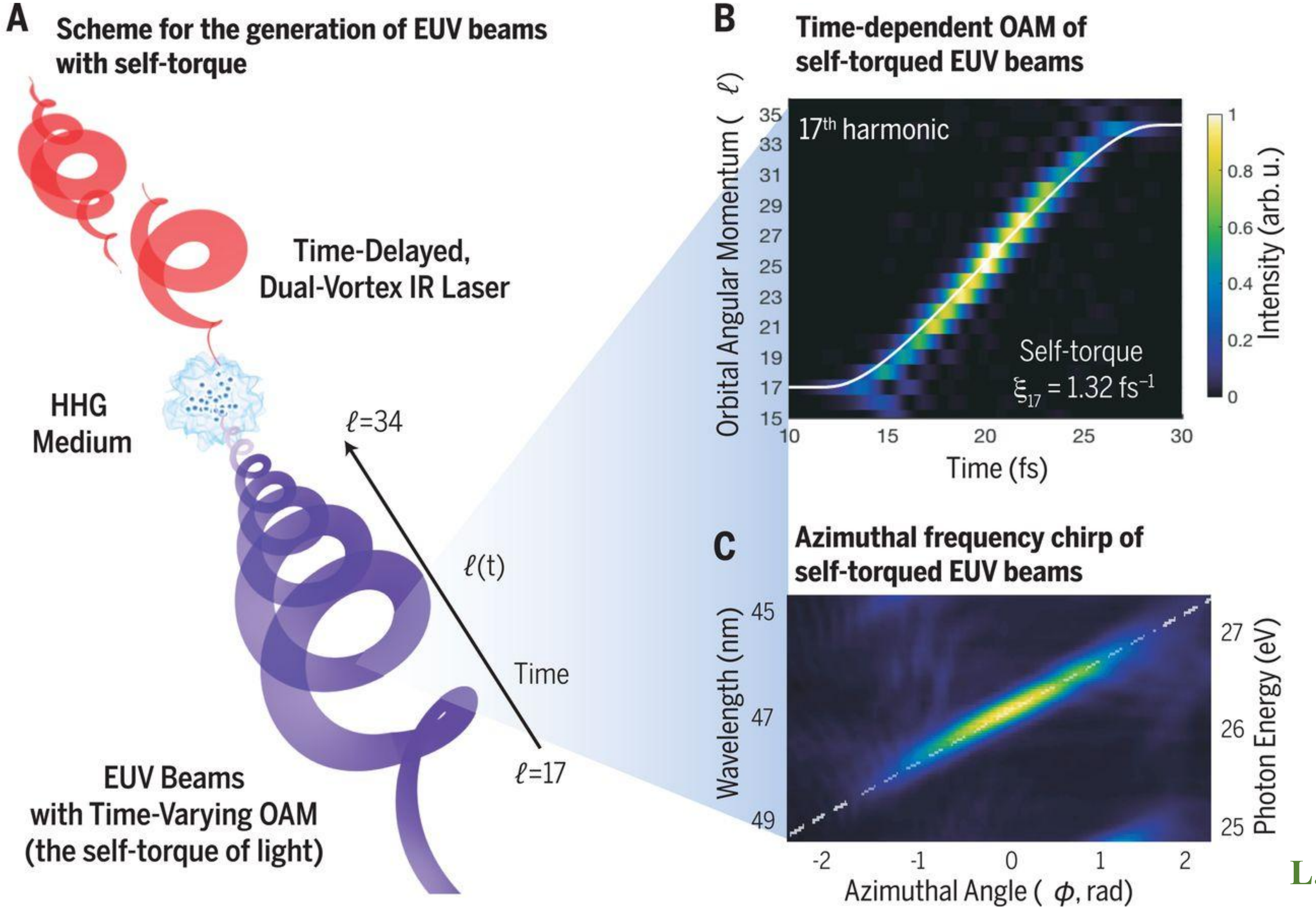

L. Rego, et al. Physical Review Letters 117, 163202 (2016).

# Generation of light with time-dependent OAM

- Introducing a time delay between the 2 driving vortex beams:



# Generation of light with time-dependent OAM




Definition of the self-torque of light:

$$\xi_q = \frac{d\ell_q(t)}{dt}$$

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M. M. Murnane

ICFO

E. Pisanty
M. Lewenstein

L. Rego, et al. Science, 364, eaaw9486 (2019)

# Generation of light with time-dependent OAM

- The definition of the self-torque of light:

$$\xi_q = \frac{d\ell_q(t)}{dt}$$

- The measurement:

$$LG_{\ell,p}(\rho,\phi,z;k_0) = E_0 \frac{w_0}{w(z)} \left(\frac{\sqrt{2}\rho}{w(z)}\right)^{|\ell|} L_p^{|\ell|}\left(\frac{2\rho^2}{w^2(z)}\right) \times \exp\left(-\frac{\rho^2}{w^2(z)}\right) \exp\left[i\ell\phi + i\frac{k_0\rho^2}{2R(z)} + i\Phi_G(z)\right]$$

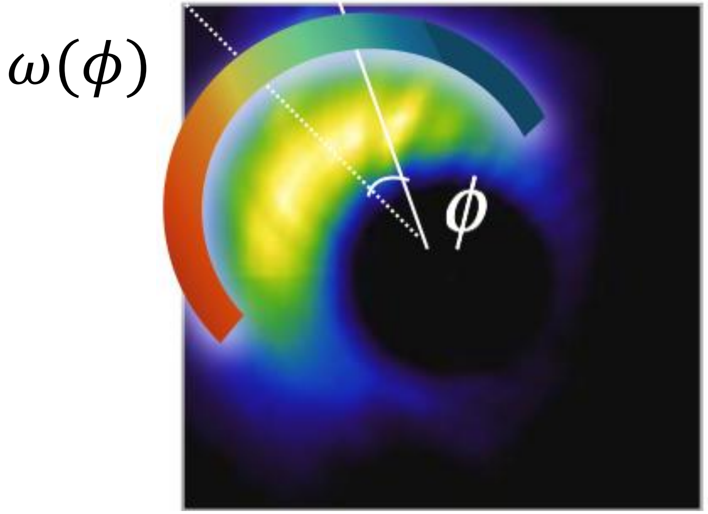
$\ell \rightarrow \ell(t)$

Instantaneous frequency:

$$\omega_q(t,\phi) = \frac{d\varphi_q(t,\phi)}{dt} = \omega_q + \frac{d\ell_q(t)}{dt}\phi \approx \omega_q + \xi_q\phi$$

*The self-torque imprints an azimuthal frequency chirp!*

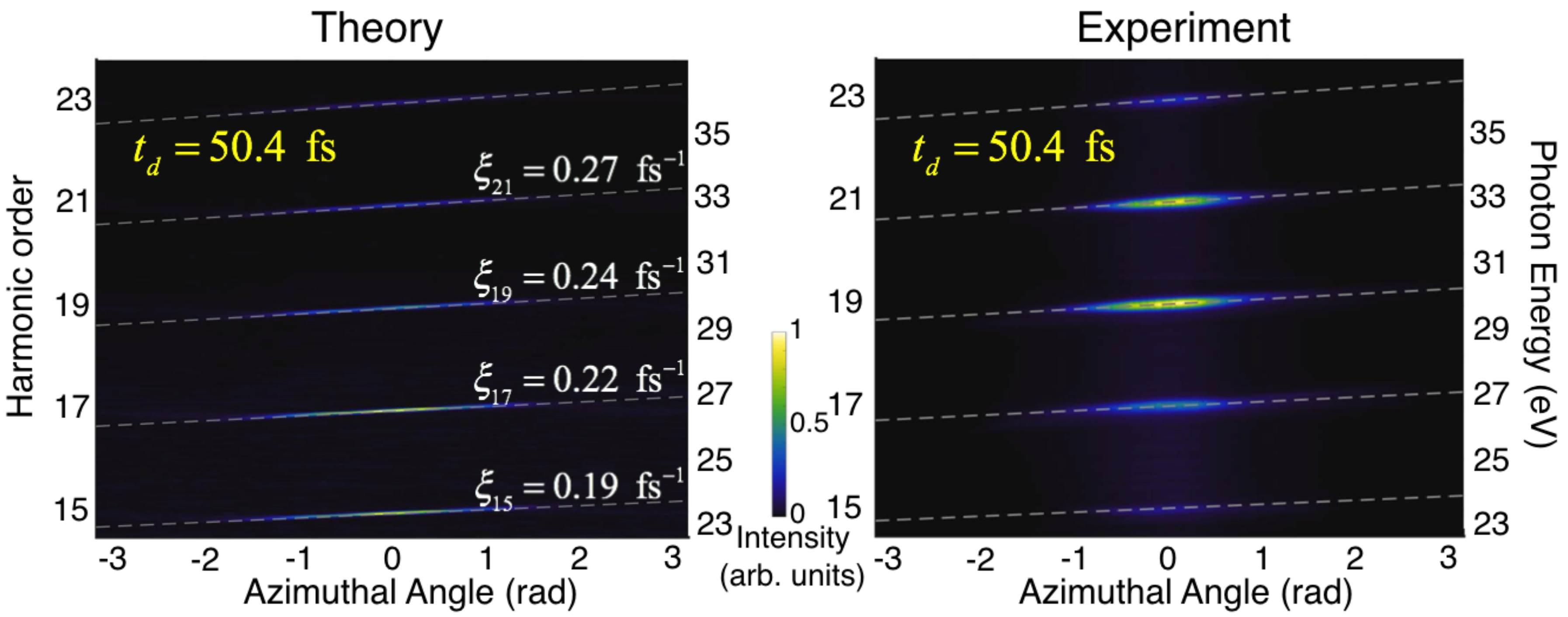
Intensity profile of the HHG beam




L. Rego, et al. Science, 364, eaaw9486 (2019)

# Generation of light with time-dependent OAM

- The experimental confirmation:




The slope is the self-torque!

$$\omega_q(t,\phi) = \frac{d\varphi_q(t,\phi)}{dt} = \omega_q + \frac{d\ell_q(t)}{dt}\phi \approx \omega_q + \xi_q \phi$$

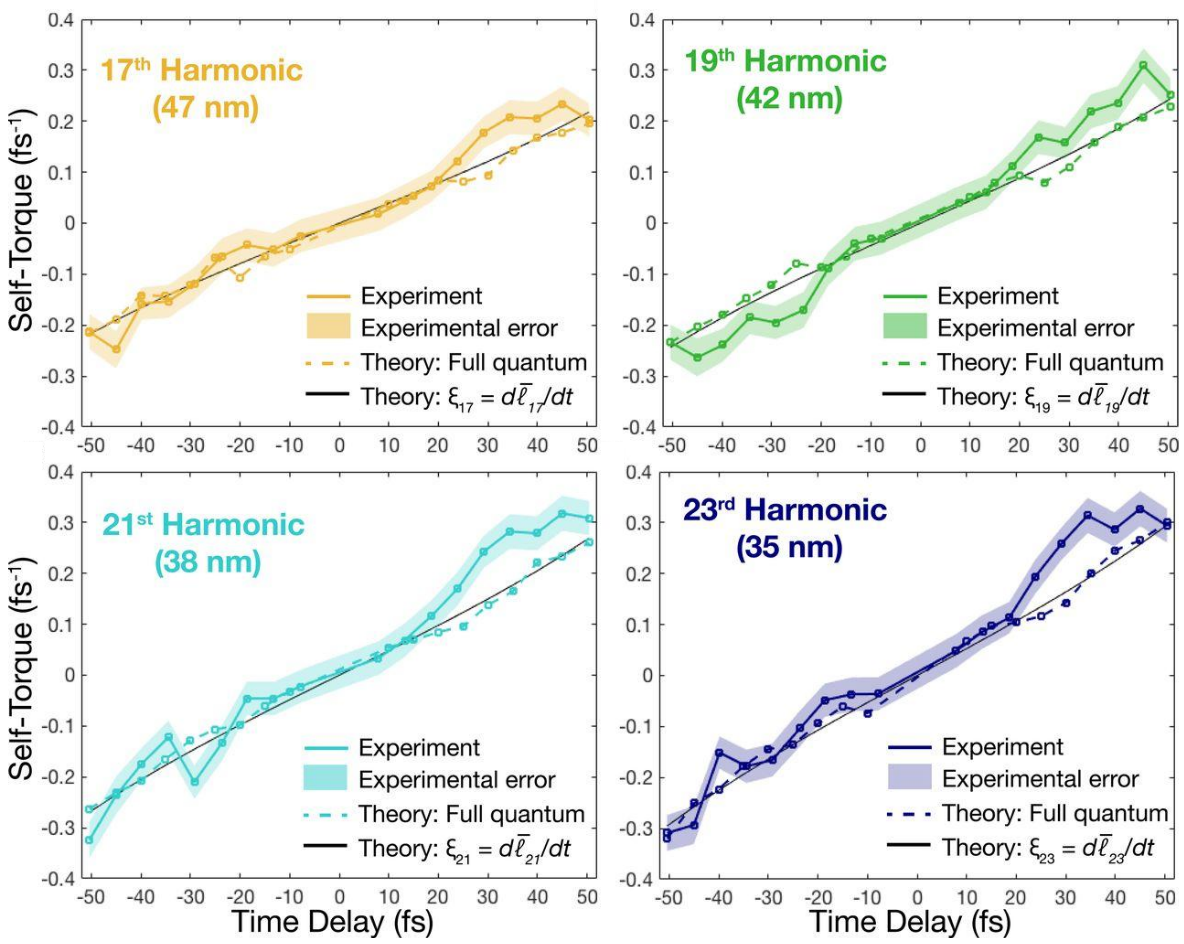
$\ell_1 = 1$ 50 cycles (50 fs FWHM)

$\ell_1 = 2$ 50 cycles (50 fs FWHM)

L. Rego, et al. Science, 364, eaaw9486 (2019)

# Generation of light with time-dependent OAM

- The control over the amount of self-torque by changing the time delay:

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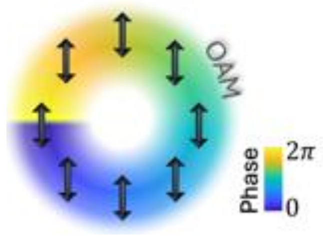
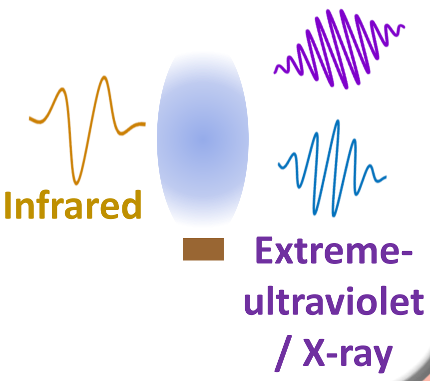
Structured light + High-order harmonic generation

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## Spectroscopy

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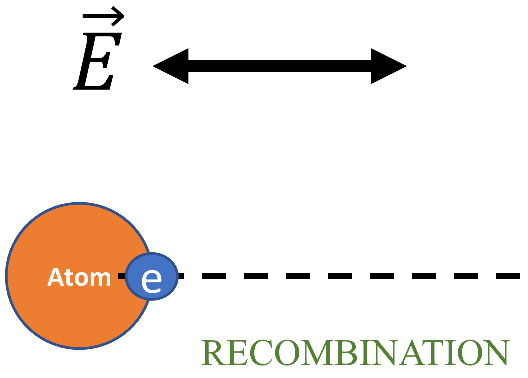
# Control of the polarization of the attosecond pulses

- The efficiency of HHG decreases greatly with the ellipticity of the driving field

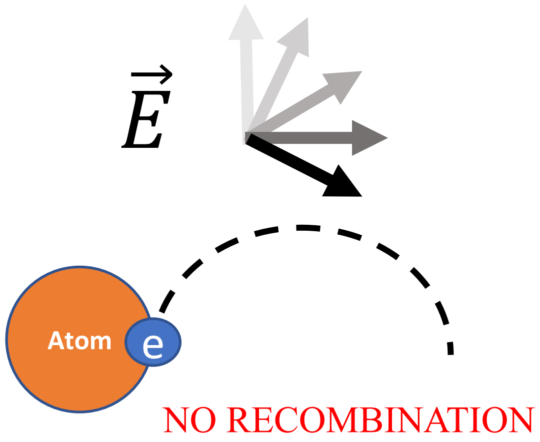
K. S. Budil, et al. Physical Review A 48, R3437(R) (1993)

**Linearly-polarized driving field**



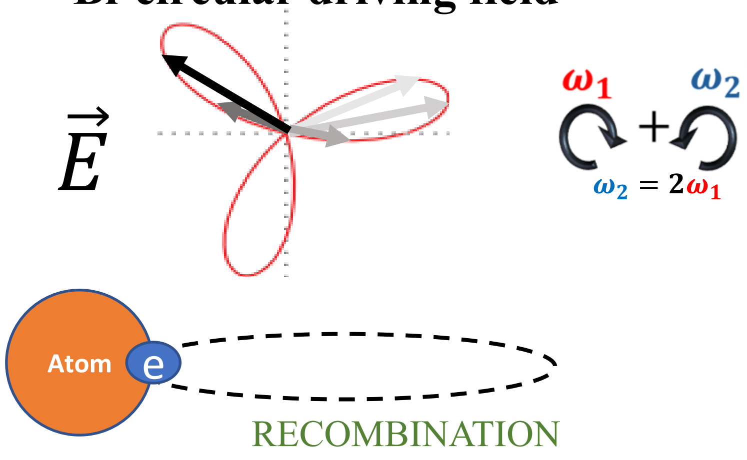

Linearly-polarized harmonics

**Circularly-polarized driving field**




No harmonics

**Bi-circular driving field**

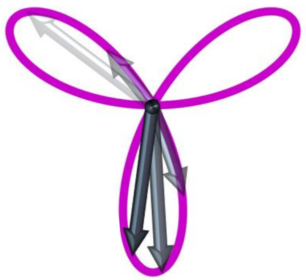



Circularly-polarized harmonics!

H. Eichmann, et al. Physical Review A, 51, R3414–R3417 (1995)
S. Long, et al. Physical Review A, 52, 2262–2278 (1995)
D. B. Miloševic, et al. Physical Review A, 61, 063403 (2000)
A. Fleischer, et al. Nature Photonics, 8, 543–549 (2014)
O. Kfir, et al. Nature Photonics, 9, 99–105 (2015)

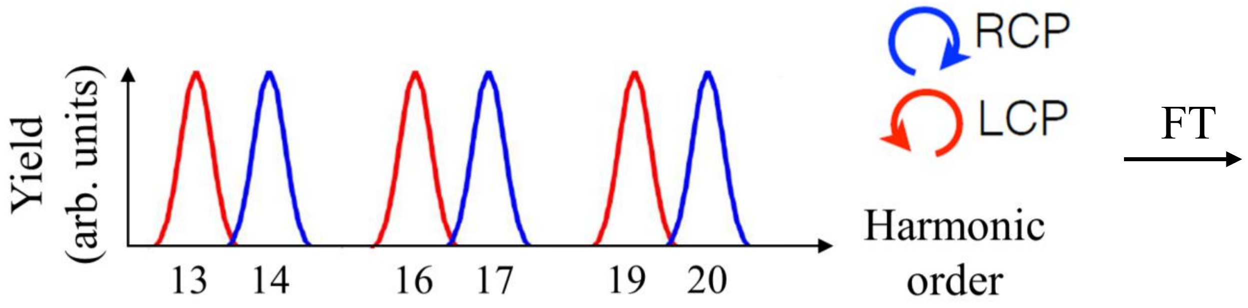
# Control of the polarization of the attosecond pulses

- The bi-circular field allows for the generation of circularly polarized harmonics



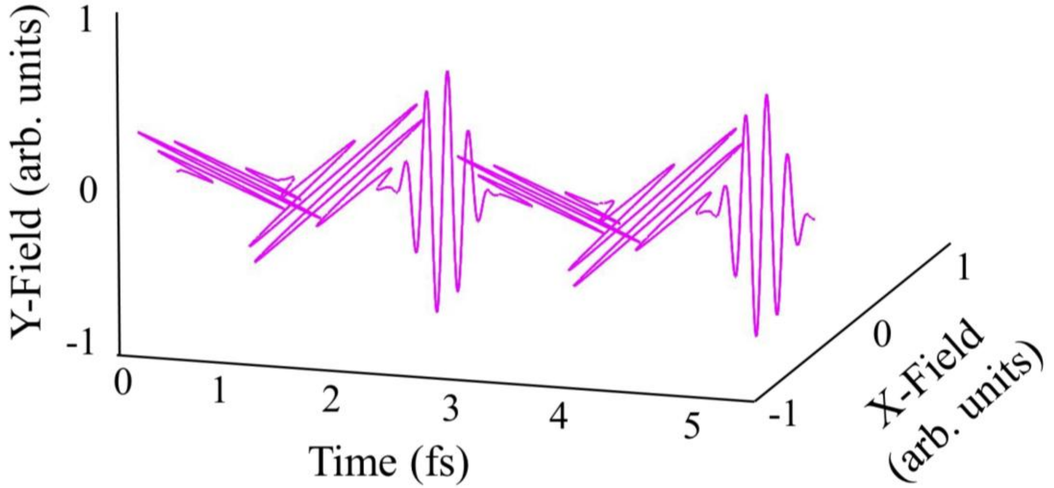
HHG spectrum: circularly polarized harmonics

Circular!



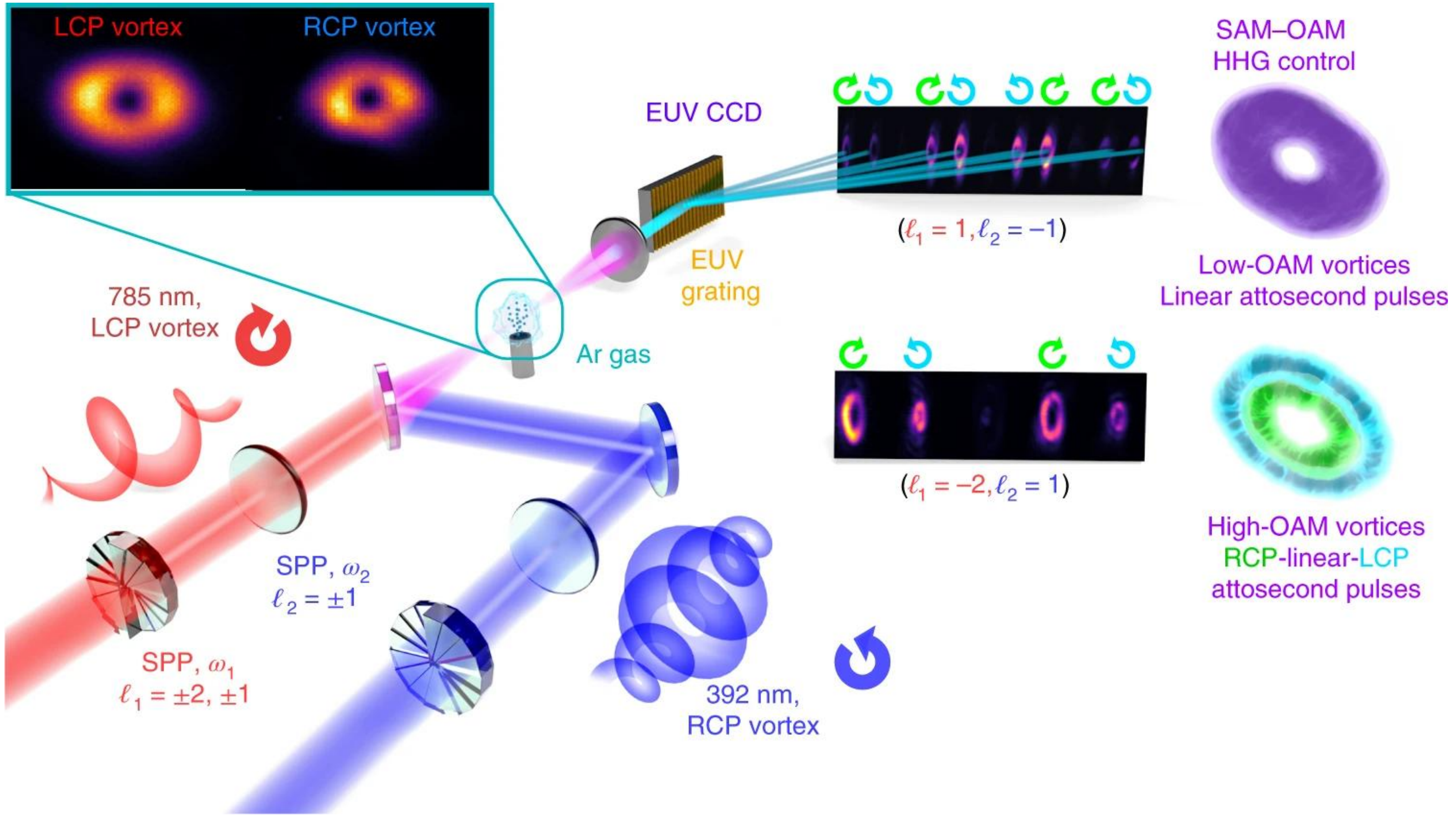

Polarization of the attosecond pulses

Still linear!



# Control of the polarization of the attosecond pulses through OAM

- Scheme of HHG driven by a bi-circular vortex field





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Q. Nguyen
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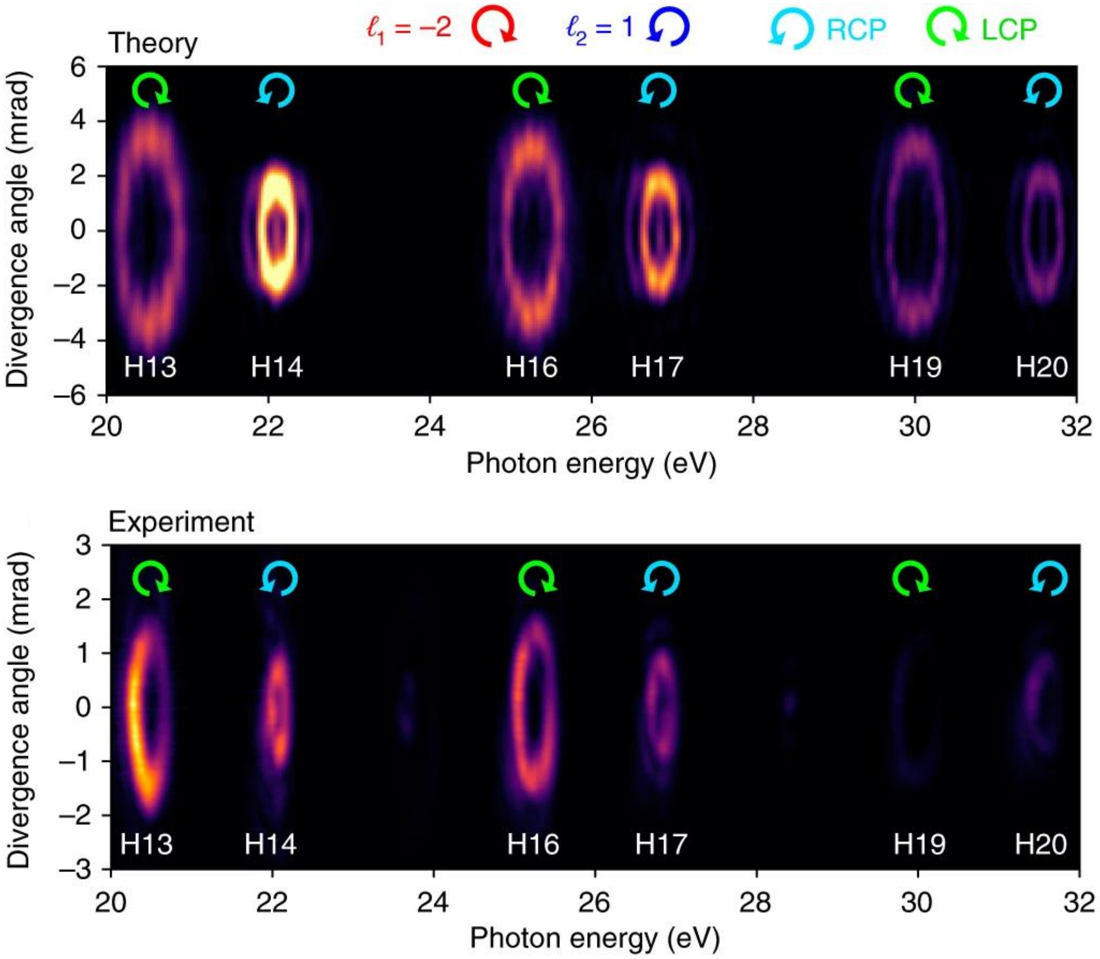
H. C. Kapteyn
M. M. Murnane


**K. M. Dorney, L. Rego, et al. Nature Photonics, 13, 123–130 (2018)**

# Control of the polarization of the attosecond pulses through OAM

- For certain OAM combinations, the harmonics with opposite helicity are spatially separated




Selection rules for the OAM of the harmonics

$$\begin{cases} \ell_q = n_1\ell_1 + n_2\ell_2 \\ \sigma_q = n_1\sigma_1 + n_2\sigma_2 \\ q\omega_1 = n_1\omega_1 + n_2\omega_2 \end{cases} \rightarrow \ell_q = \frac{q + 2\sigma_q\sigma_1}{3}(\ell_1 + \ell_2) - \sigma_q\sigma_1\ell_2$$

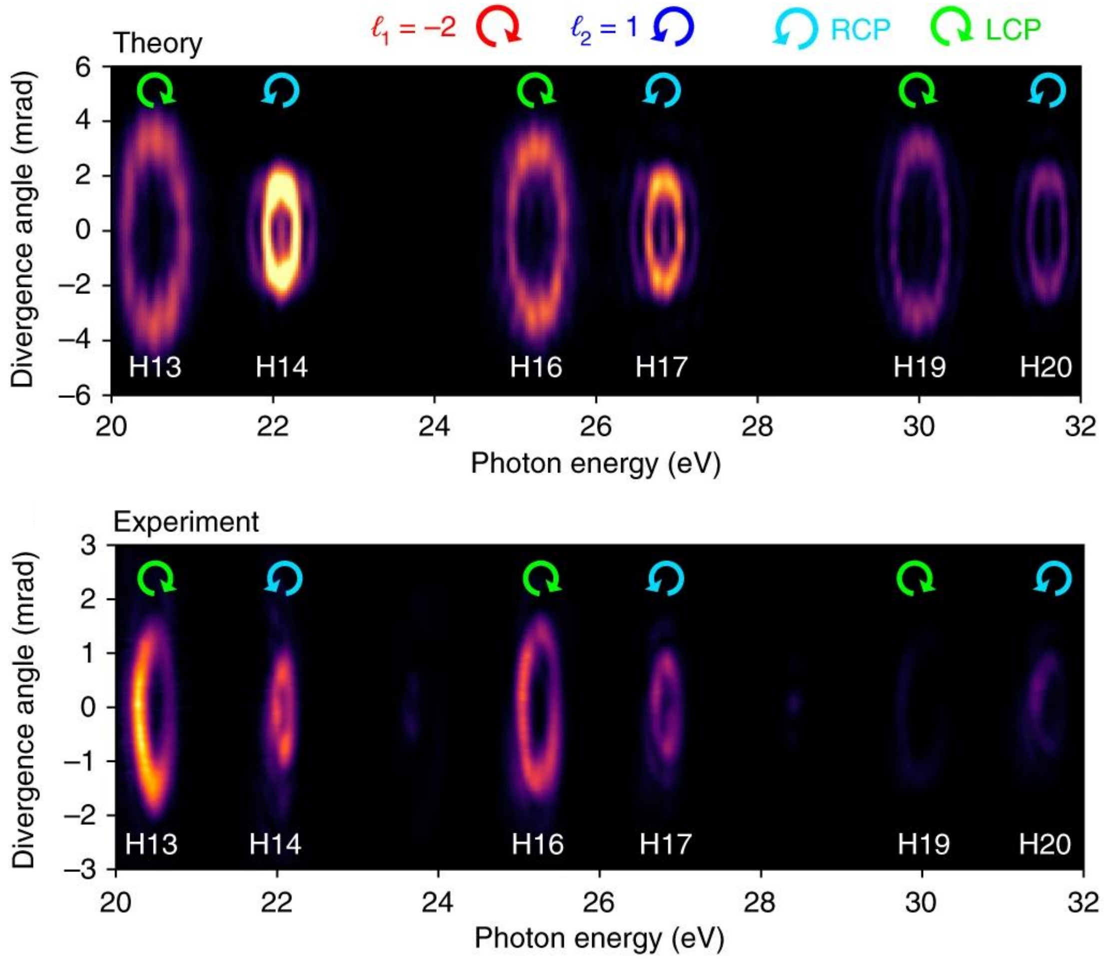
Different OAM ⟺ Different divergence

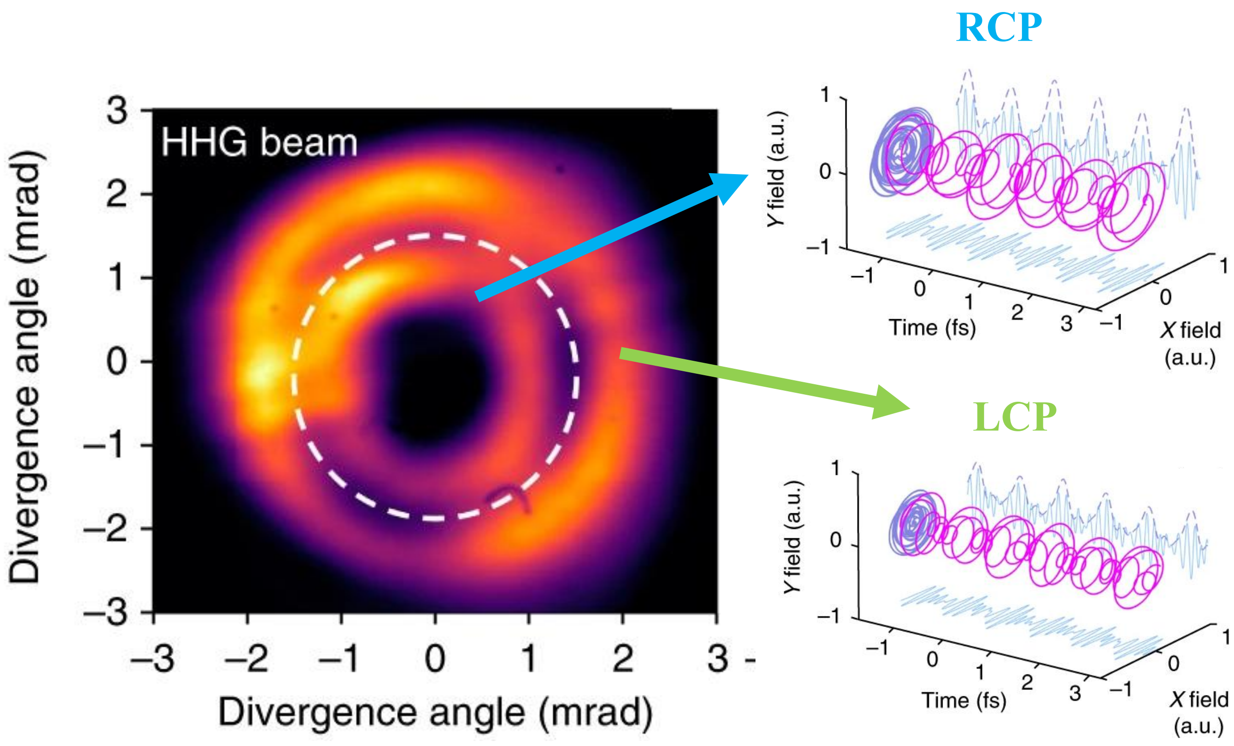
Spatial separation of harmonics
with opposite polarization!

K. M. Dorney, L. Rego, et al. Nature Photonics, 13, 123–130 (2018)

# Control of the polarization of the attosecond pulses through OAM

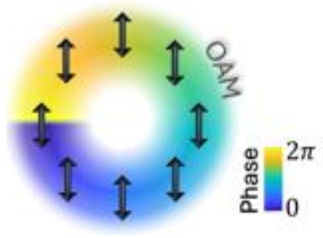
- For certain OAM combinations, the harmonics with opposite helicity are spatially separated

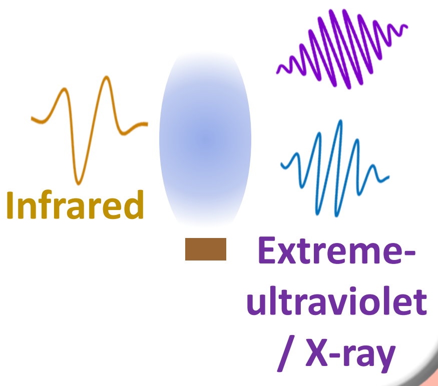



K. M. Dorney, L. Rego, et al. Nature Photonics, 13, 123–130 (2018)

Structured light + High-order harmonic generation




## Control over emitted light

=

- ❑ Extreme-ultraviolet / X-ray structured light
- ❑ New types of structured light: self-torque of light

=

- ❑ Control over other properties of the extreme-ultraviolet/X-ray light: polarization or spectral content.

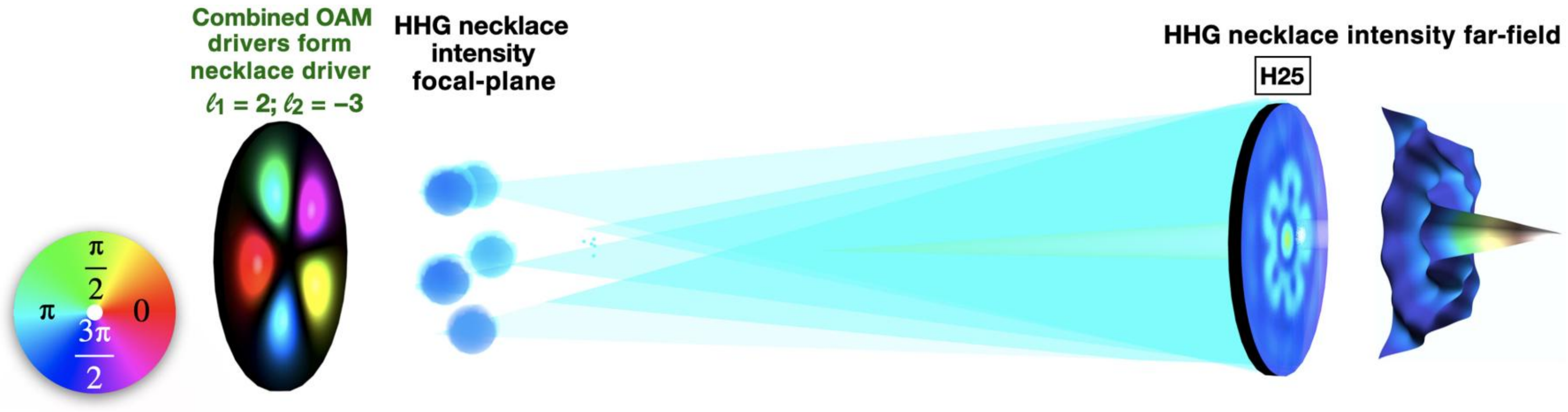
## Spectroscopy

=

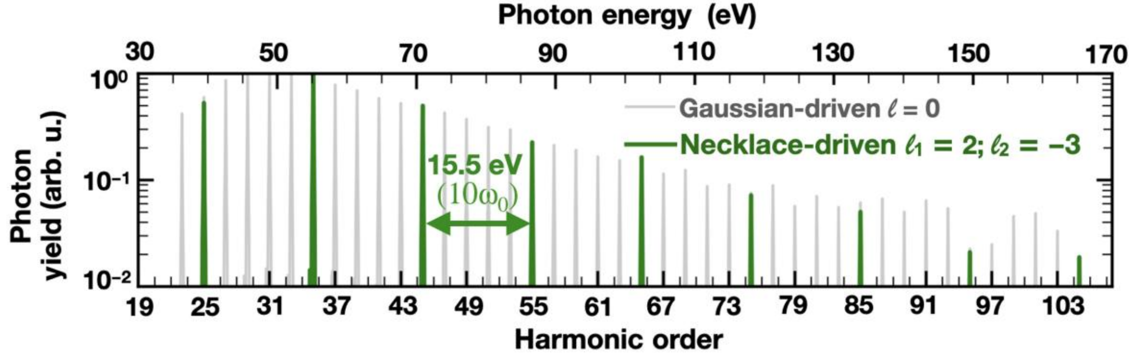
- ❑ Information about the target: chirality

# Control of the harmonic comb through OAM

- HHG driven by a necklace beam (combination of two vortex beams): only some harmonics interfere constructively on-axis




*5 time-delayed emitters*





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K. M. Dorney
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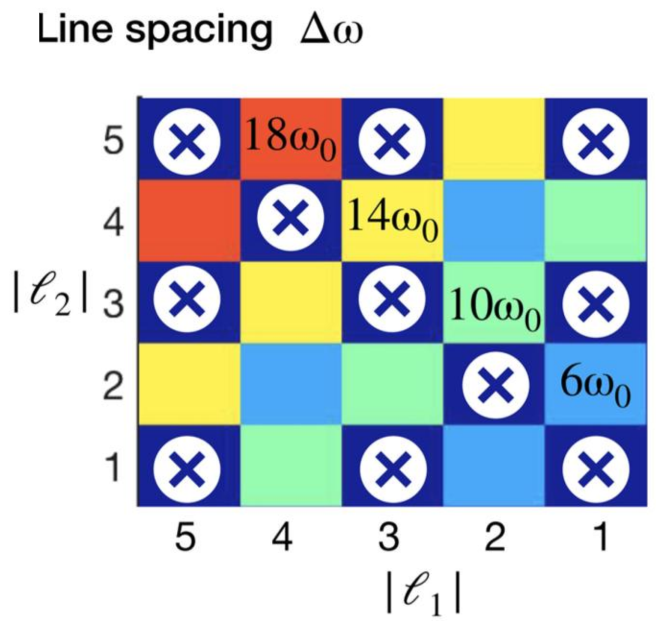
I. Binnie
H. C. Kapteyn
M. M. Murnane

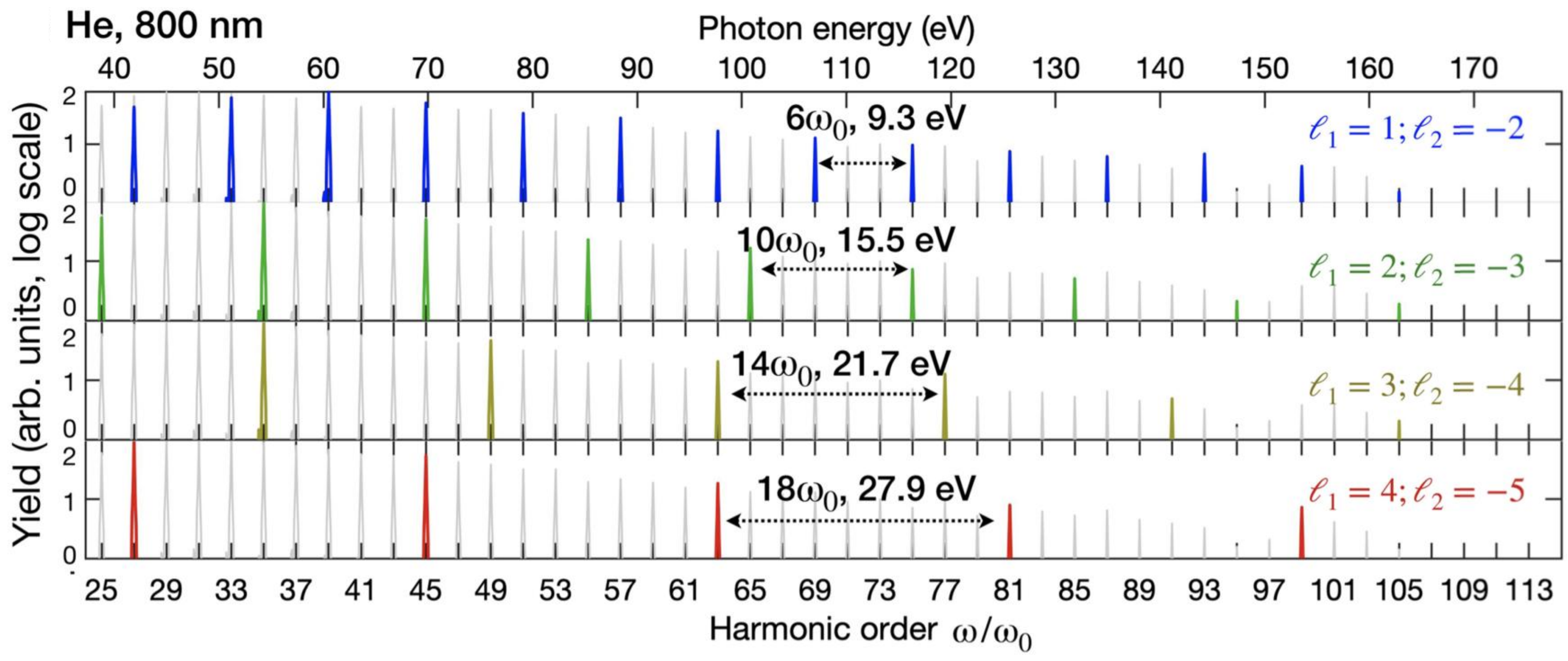


**L. Rego, N. J. Brooks, Q. L. D. Nguyen, et al. Science Advances. 8, eabj7380 (2022).**

# Control of the harmonic comb through OAM

- Control over the line-spacing in the harmonic combs through the driving's OAM

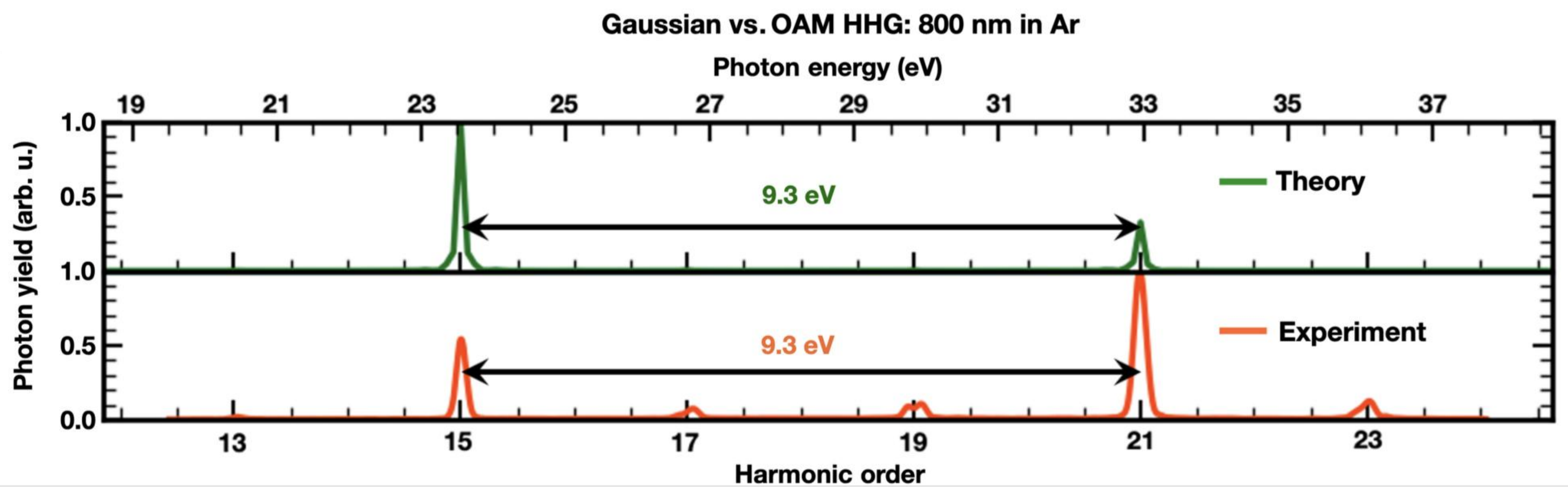



L. Rego, N. J. Brooks, Q. L. D. Nguyen, et al. Science Advances. 8, eabj7380 (2022).

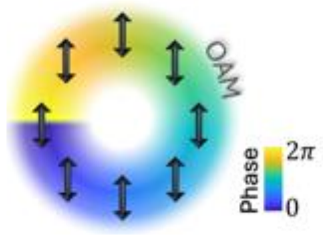
# Control of the harmonic comb through OAM

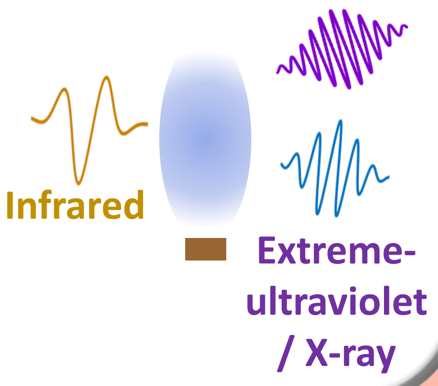
- ❑ The experimental demonstration




L. Rego, N. J. Brooks, Q. L. D. Nguyen, et al. Science Advances. 8, eabj7380 (2022).

Structured light + High-order harmonic generation




## Control over emitted light

= 
- ❑ Extreme-ultraviolet / X-ray structured light
- ❑ New types of structured light: self-torque of light

= 
- ❑ Control over other properties of the extreme-ultraviolet/X-ray light: polarization or spectral content.

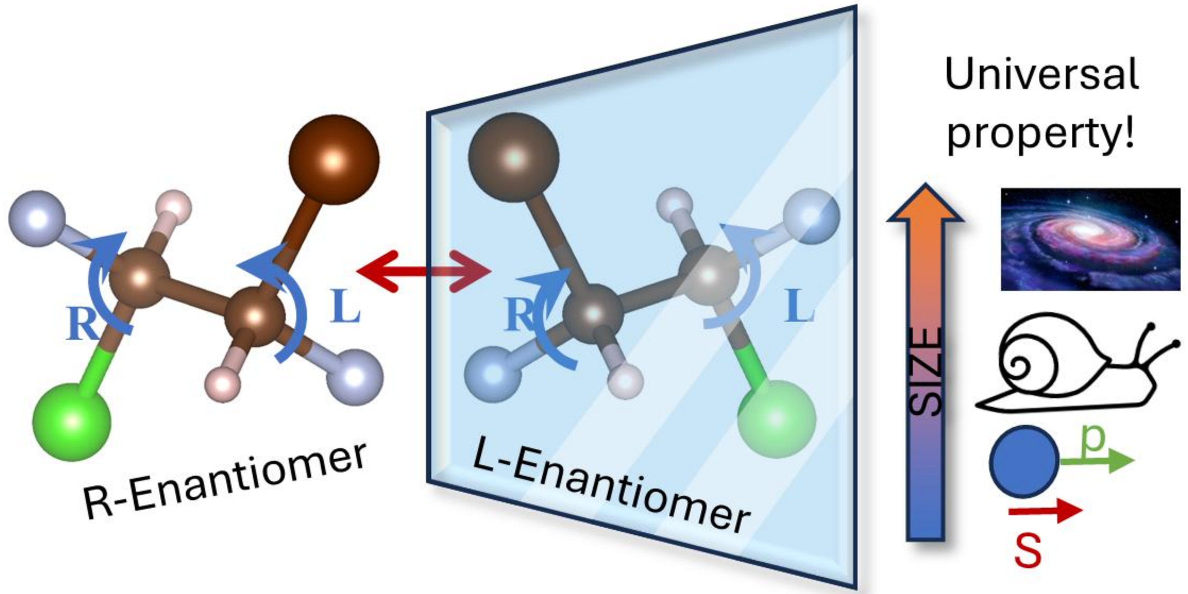
## Spectroscopy

= 
- ❑ Information about the target: chirality

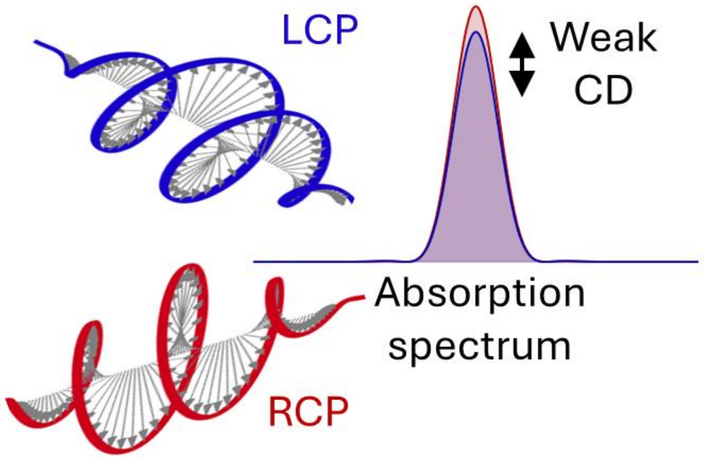
# Chirality and Circular Dichroism

- Chiral objects cannot be superimposed onto their mirror image.
- The two versions of a chiral molecule are called enantiomers.
- The distinction between enantiomers is vital in pharmaceutical industry.

The concept of chirality




Standard CD




Electric+magnetic dipole interactions

D. Habibović, K. Hamilton, O. Neufeld & L. Rego. Nat Rev Phys 6, 663–675 (2024)

# Ultrafast and topological chiral sensing with vector beams

- HHG driven by a vector beam in chiral molecules

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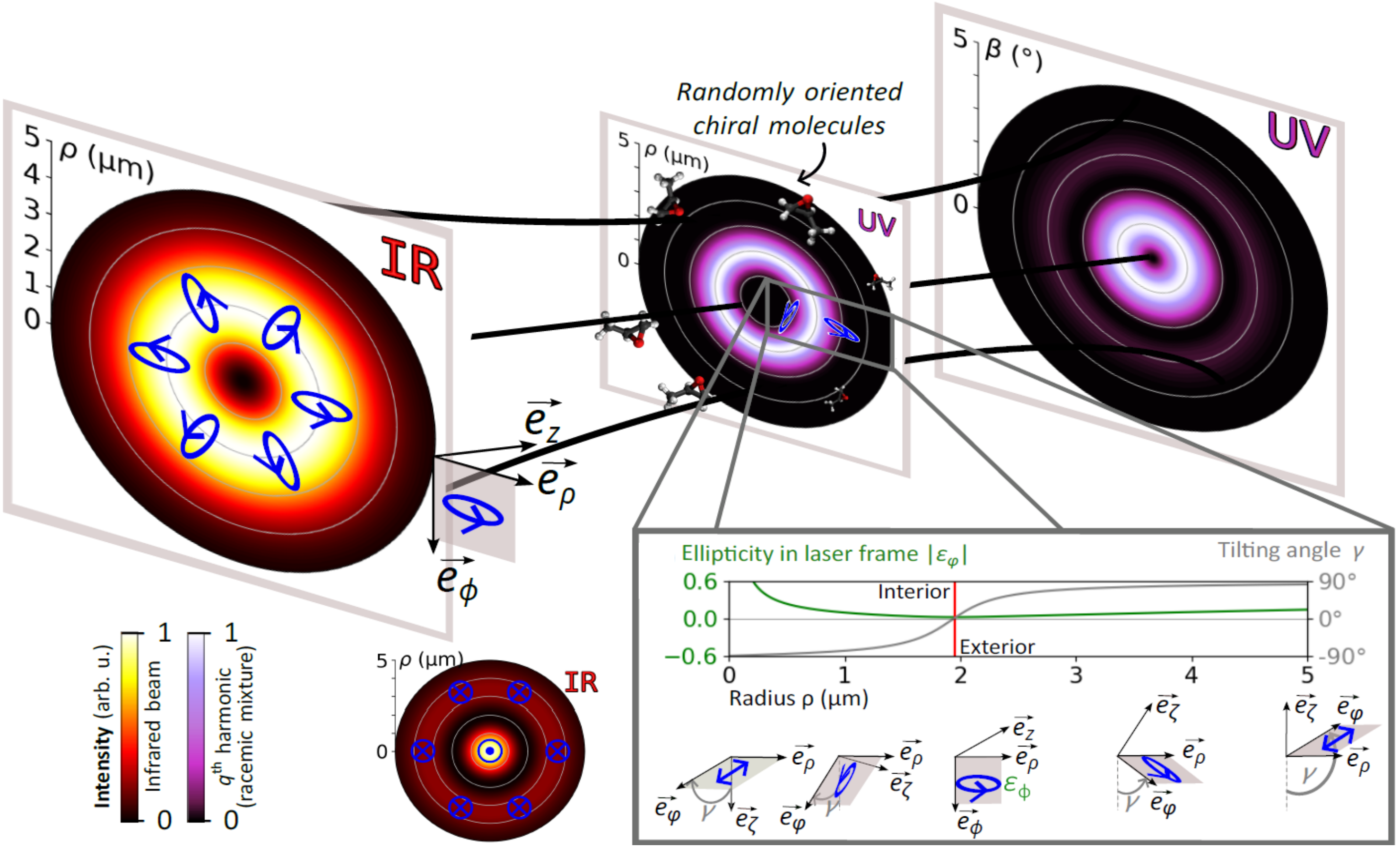
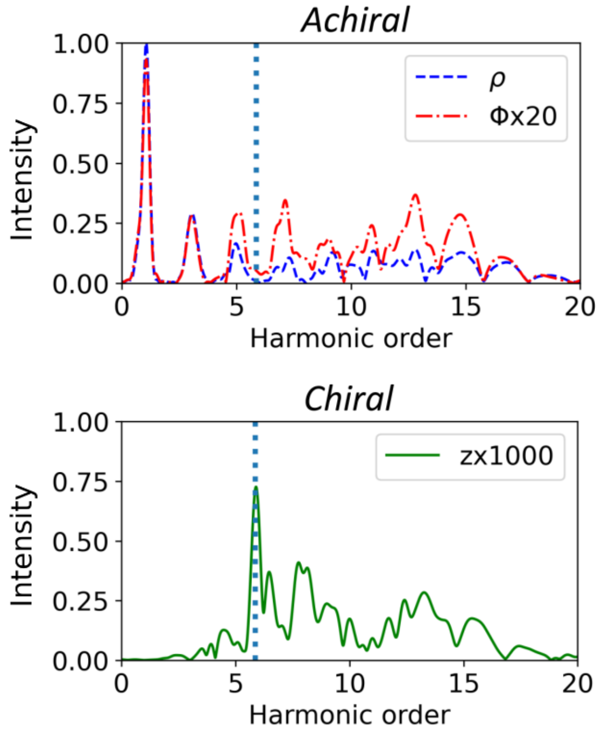
A. Rodriguez

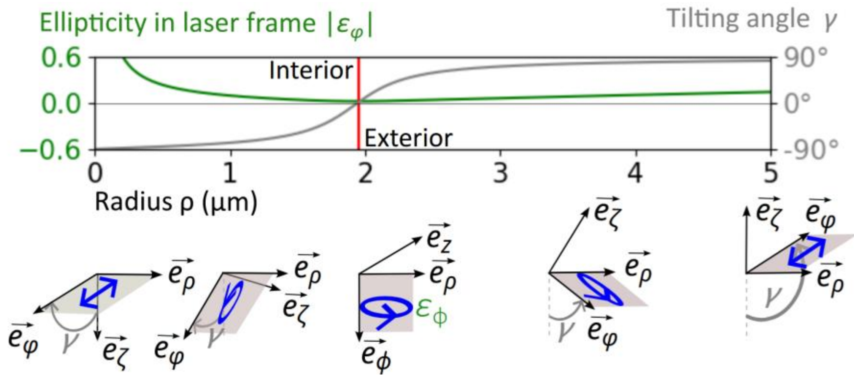



Stronger chiral component in the highly nonlinear regime! But it is parallel to the propagation direction

**A. Rodriguez & L. Rego. ArXiv2606:13402v1. Under review (2026)**

# Ultrafast and topological chiral sensing with vector beams




Rotation of the laser polarization frame (due to tight-focusing)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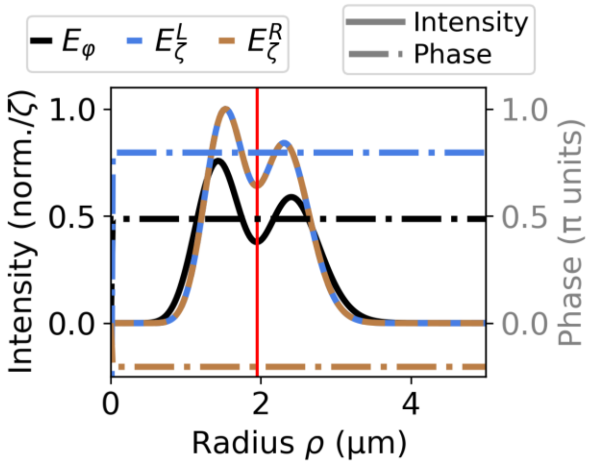
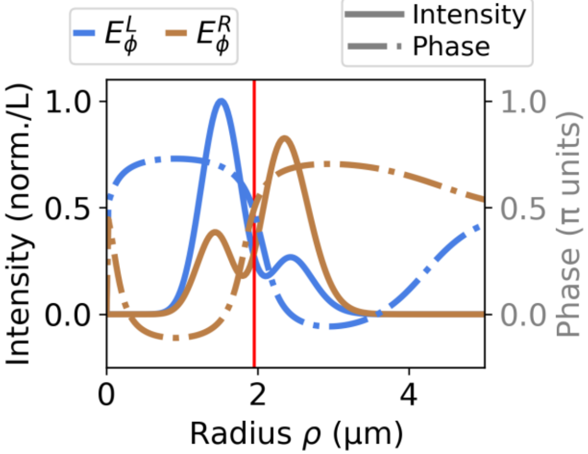
A. Rodriguez


Far-field intensity profiles of the 6th harmonic

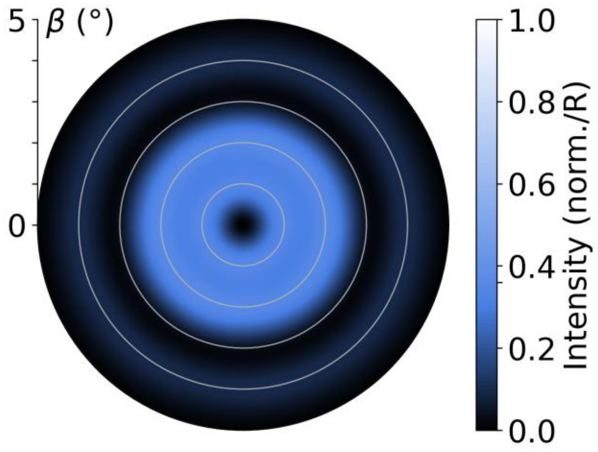
Laser frame



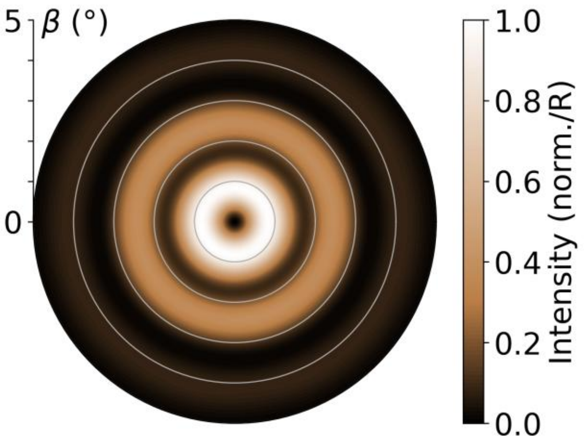

Laboratory frame:
interference between chiral and achiral emissions




L-enantiomer




R-enantiomer




Achiral

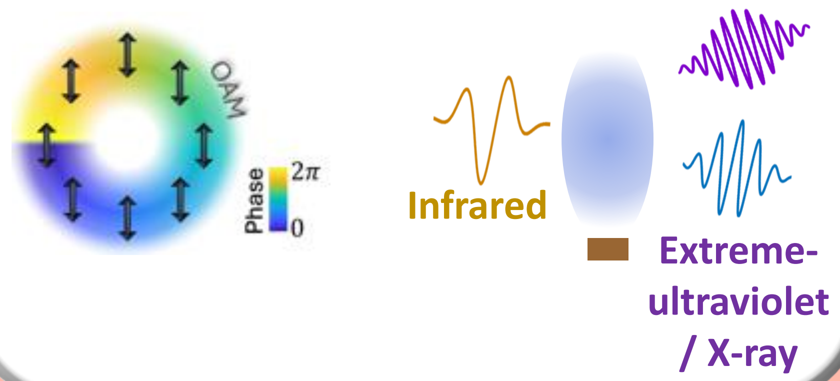
L-enantiomer

R-enantiomer

A. Rodriguez & L. Rego. ArXiv2606:13402v1. Under review (2026)

# Conclusions

Structured light + High-order harmonic generation




### Control over emitted light

=
- ❑ Extreme-ultraviolet / X-ray structured light
- ❑ New types of structured light: self-torque of light

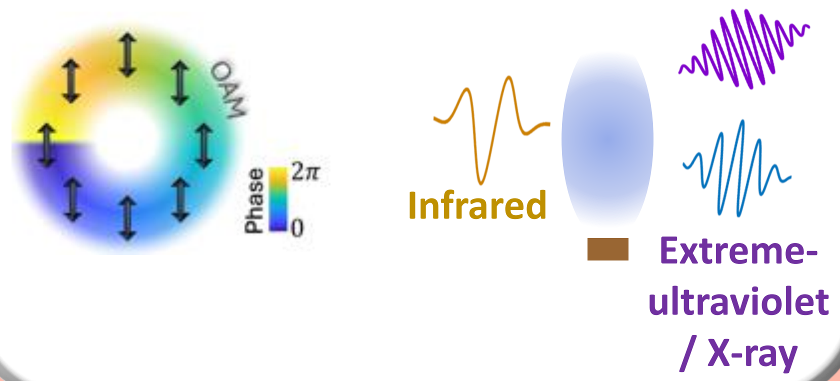
=
- ❑ Control over other properties of the extreme-ultraviolet/X-ray light: polarization or spectral content.

### Spectroscopy

=
- ❑ Information about the target: chirality

# Conclusions

**Structured light** + **High-order harmonic generation**




**Control over emitted light**

= 
- ❑ **Extreme-ultraviolet / X-ray structured light**
- ❑ **New types of structured light: self-torque of light**

Why? → Because HHG is highly nonlinear

=
- ❑ **Control over other properties of the extreme-ultraviolet/X-ray light: polarization or spectral content.**

**Spectroscopy**

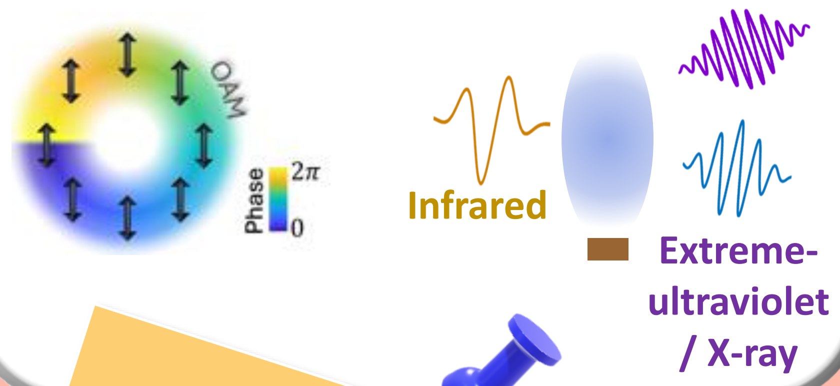
=
- ❑ **Information about the target: chirality**

Why? → Why? →

Structured light
↓
Control over spatial distribution
↓
- ❑ Decompose light
- ❑ Create interferences to suppress or enhance emissions

# Conclusions

**Structured light** + **High-order harmonic generation**




**Structured light is a powerful control knob for ultrafast physics**

**Control over emitted light**

= 
- ❑ **Extreme-ultraviolet / X-ray structured light**
- ❑ **New types of structured light: self-torque of light**

Why? → Because HHG is highly nonlinear

= 
- ❑ **Control over other properties of the extreme-ultraviolet/X-ray light: polarization or spectral content.**

**Spectroscopy**

= 
- ❑ **Information about the target: chirality**

Why? → Structured light → Control over spatial distribution →
- ❑ Decompose light
- ❑ Create interferences to suppress or enhance emissions

# Acknowledgements

VNiVERSiDAD D SALAMANCA

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University of Colorado Boulder

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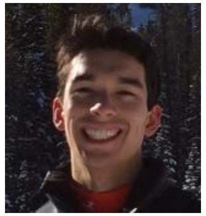

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Quynh Nguyen

UAM Universidad Autónoma de Madrid

IMPERIAL

**Other collaborators and colleagues:**

| | | |
|---|---|---|
| David Ayuso | Anne L'Huillier | Alex Turpin |
| Jon Marangos | Olga Smirnova | Alicia Palacios |
| Fernando Martín | Ming-Chang Chen | Jesús González |

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# Thank you very much!

# BACKUP SLIDES

# Macroscopic theoretical models

- Full Quantum Strong Field Approximation (SFA) + Propagation

-Full Quantum SFA (no saddle point approximation) for the computation of the elementary radiators.

Hamiltonian

$$H(t) = \frac{p^2}{2m} + V_C(\mathbf{r}) + V_F(\mathbf{P}, t)$$

Evolution of the wavefunction

$$|\delta\psi(t)\rangle = \frac{\mathrm{i}}{\hbar}\int_{t_0}^{t} G_F(t, t_1)V_F(t_1)G_0(t_1, t_0)|\phi_0\rangle \mathrm{d}t_1$$

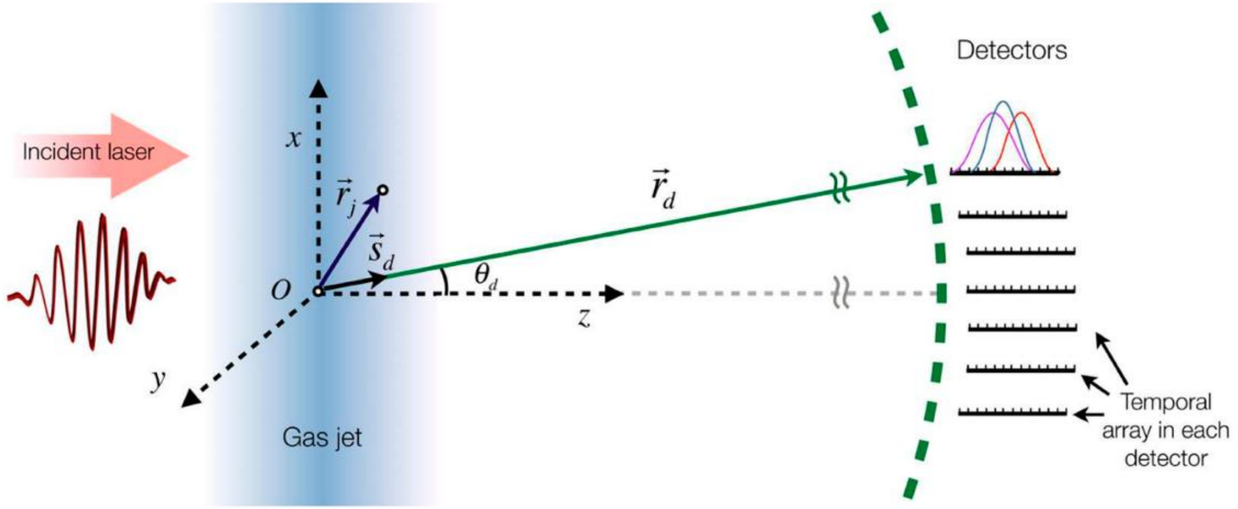
Dipolar acceleration

$$\mathbf{a}(\mathbf{P}, t) = (1/m)\langle\phi_0(t)|(-\nabla V_C)|\mathbf{P}\rangle\delta\psi(\mathbf{P}, t)$$

J. A. Pérez-Hernández, et al. Optics Express 17, 9891 (2009)

-Harmonic propagation: electromagnetic field propagator.

$$\mathbf{E}_i^j(\mathbf{r}_d, t) = \frac{q_j}{c^2|\mathbf{r}_d - \mathbf{r}_j(0)|}\mathbf{s}_d \times \{\mathbf{s}_d \times \mathbf{a}_j[t - |\mathbf{r}_d - \mathbf{r}_j(0)|/c]\}$$




C. Hernández-García, et al., Physical Review A 82, 033432 (2010)

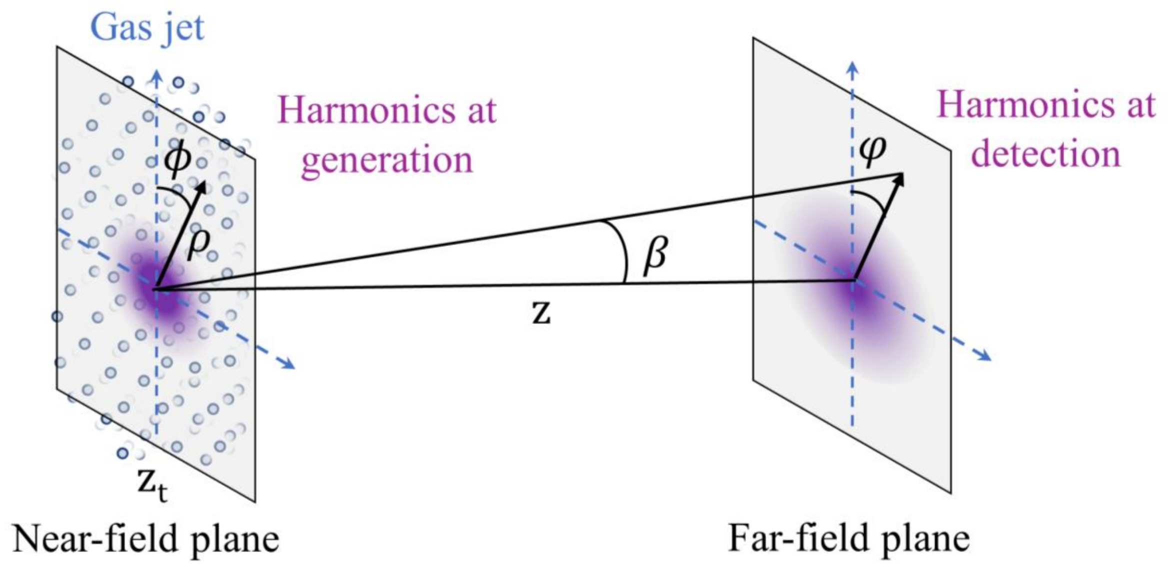
# Macroscopic theoretical models

❑ Thin Slab Model

Driving field: $U(\rho, \phi, z_t)$

Harmonic field:

$$A_q(\rho, \phi) \propto \sum_r |U(\rho, \phi, z_t)|^p e^{iq\Phi(\rho,\phi,z_t)} e^{i\alpha_q^r |U(\rho,\phi,z_t)|^2}$$




Far field using Fraunhofer diffraction:

$$U_q(\beta, \varphi, z) = -iq \frac{e^{ikz} e^{i\frac{kz\tan^2\varphi}{2}}}{z\lambda} \int_0^\infty \int_0^{2\pi} A_q(\rho, \phi) e^{-i\frac{2\pi}{\lambda} qr \tan\beta \cos(\varphi-\phi)} \rho d\rho d\phi$$

- Fast calculations
- To distinguish the role of the nonperturbative parameters (dipole phase and amplitude scaling power)

C. Hernández-García, et al. New Journal of Physics 17, 093029 (2015)

# Theoretical methods

## 1. Microscopic

(at one point of space, within dipole approximation)

- ❑ Single-molecule response to the local electric field: Time-Dependent Density Functional Theory (TDDFT) using the Octopus software to obtain the dipolar acceleration

Marques, M. A. et al. Comput. Phys. Commun. 151, 60–78 (2003)

- ❑ Orientational averaging (Lebedev quadrature + rotations)
- ❑ Spectrum of the light emission from the dipolar acceleration (Larmor formula)

$$E(\omega) \propto |a(\omega)|^2$$

## 2. Macroscopic

(spatially-dependent emission)

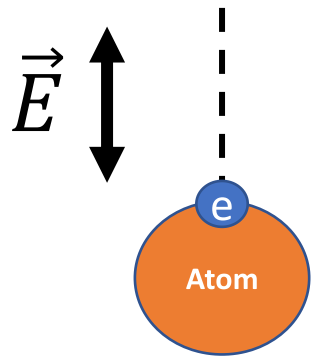
- ❑ Near field: perturbative scaling of the light emission

$$P_i = \epsilon_0 \left( \sum_j \chi_{ij}^{(1)} E_j + \sum_{jk} \chi_{ijk}^{(2)} E_{jk} + \sum_{jkl} \chi_{ijkl}^{(3)} E_{jkl} \dots \right)$$

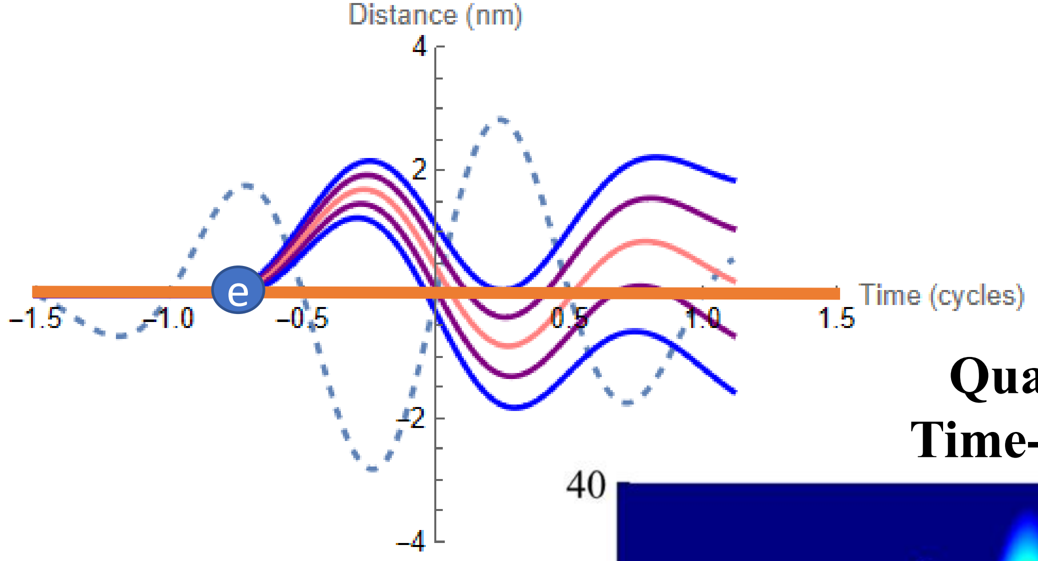
- ❑ Far field: propagation of the near field using Fraunhofer diffraction

$$E\,(k) \propto \int_{-\infty}^{\infty} E_{\perp}(x)\, e^{-i3\frac{2\pi}{\lambda_0}\sin(k)} dx$$

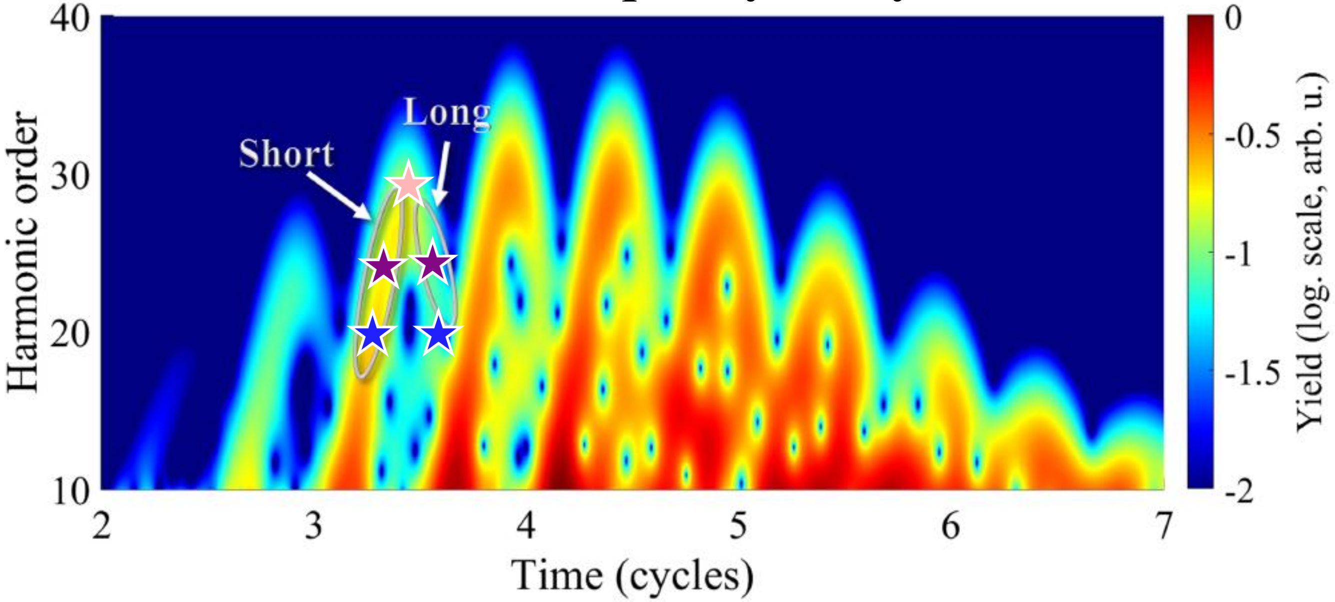
# Electron trajectories in HHG




Dynamics of the electron




**Quantum Simulations**
**Time-Frequency Analysis**



# Measurement of the attosecond pulses

- The next requirement was to develop a metrology to measure the duration of attosecond pulses and the use of suitable laser systems.

  P. Antoine, A. L'Huillier and M. Lewenstein, Phys. Rev. Lett. 77, 1234 (1996).
  P. Salières, A. L'Huillier, P. Antoine and M. Lewenstein, arXiv quant-ph/9710060 (1997).
  M. Bellini, C. Lyngå, A. Tozzi, M.B. Gaarde, T.W. Hänsch, A. L'Huillier, and C.-G. Wahlström, Phys. Rev. Lett. 81, 297 (1998).

- Prof. Agostini and co-workers had been investigating strong field phenomena and they discovered the Above Threshold Ionization

  P. Agostini, F. Fabre, G. Mainfray, G. Petite and N.K. Rahman, Phys. Rev. Lett. 42, 1127 (1979).

- One important step was taken by Prof. Agostini et al in 1994 where they investigated the principle of frequency modulation in a two-colour photon field.

  J.M. Schins, P. Breger, P. Agostini, R.C. Constantinescu, H.G. Muller, G. Grillon, A. Antonetti and A. Mysyrowicz, Phys. Rev. Lett. 73, 2180 (1994).

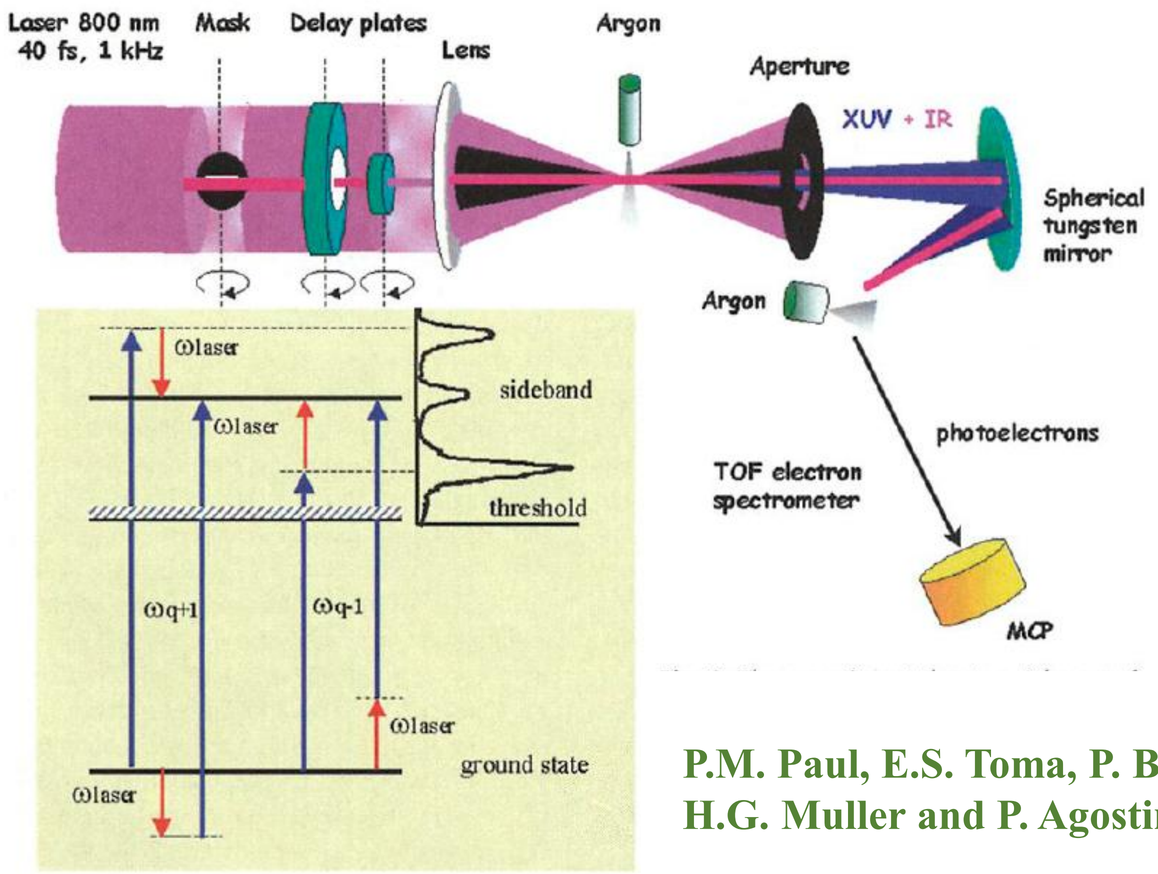
- This concept led the RABBIT (reconstruction of attosecond beating by interference of two-photon transitions) technique

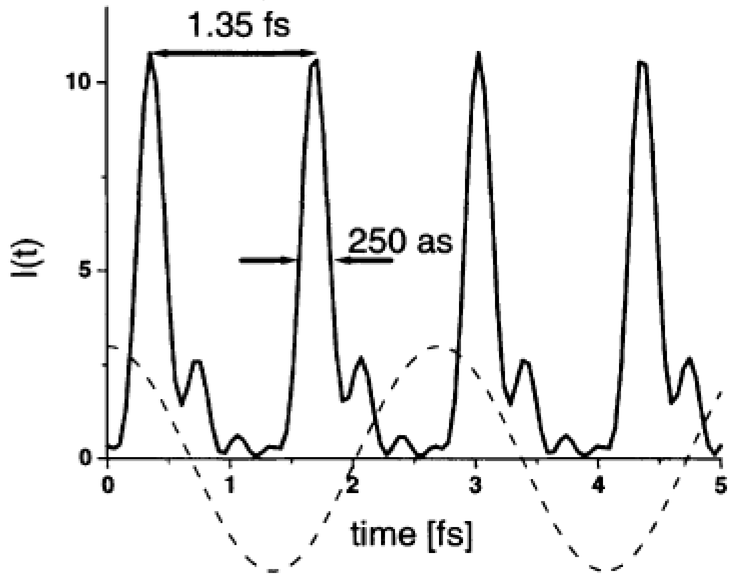
**Nobel Prize winner**

Prof. Pierre Agostini

# Measurement of the attosecond pulses

- In 2001, Prof. Agostini and co-workers measured a train of attosecond pulses for the first time using the RABBIT technique



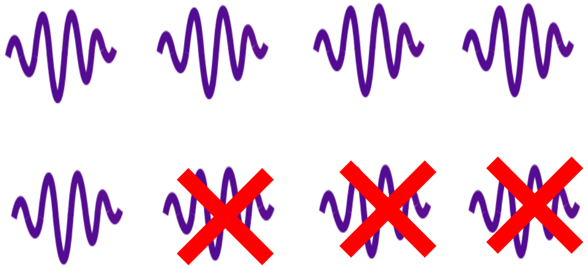

P.M. Paul, E.S. Toma, P. Breger, G. Mullot, F. Augé, Ph. Balcou, H.G. Muller and P. Agostini, Science 292, 1689 (2001).

**Nobel Prize winner**

Prof. Pierre Agostini

# Measurement of the attosecond pulses

- At the same time… there was the question of how to generate **isolated** attosecond pulses

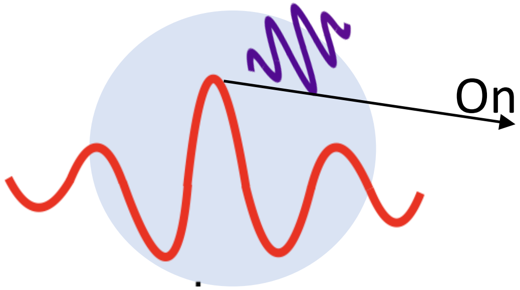


Attosecond pulse train

Isolated attosecond pulse

- Prof. Krausz and co-workers developed more advanced techniques, including the use of hollow-core fibers, to generate few-cycle visible pulses to drive HHG and achieve isolated attosecond pulses

**Ch. Spielmann, N.H. Burnett, S. Sartania, R. Koppitsch, M. Schnürer, C. Kan, M. Lenzner, P. Wobrauschek and F. Krausz, Science 278, 661 (1997).**



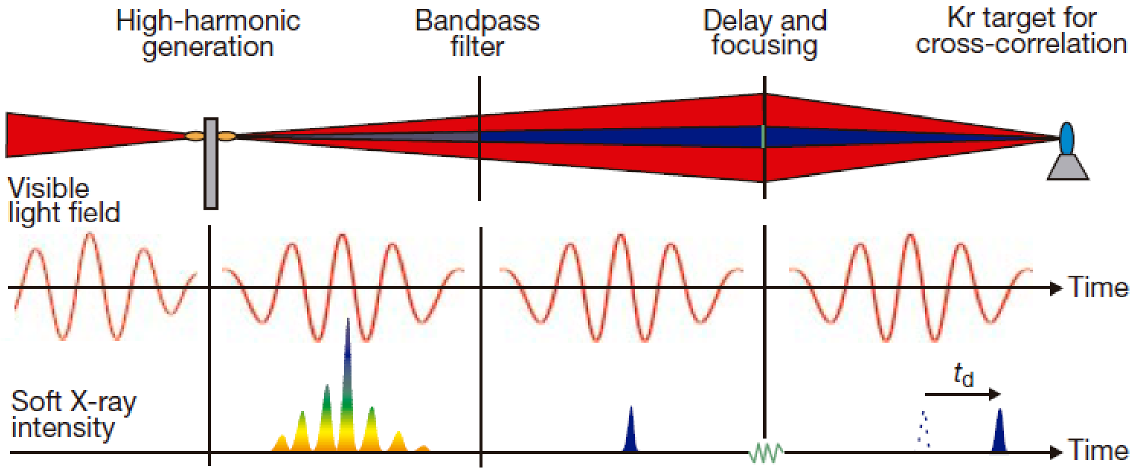
Only this semicycle is intense enough to generate very high-order harmonics

**Nobel Prize winner**

Prof. Ferenc Krausz

# Measurement of the attosecond pulses

- In 2001, Prof. Krausz and co-workers generated the first isolated attosecond pulses




M. Hentschel, R. Kienberger, Ch. Spielmann, G.A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher and F. Krausz, 414, 509 (2001).

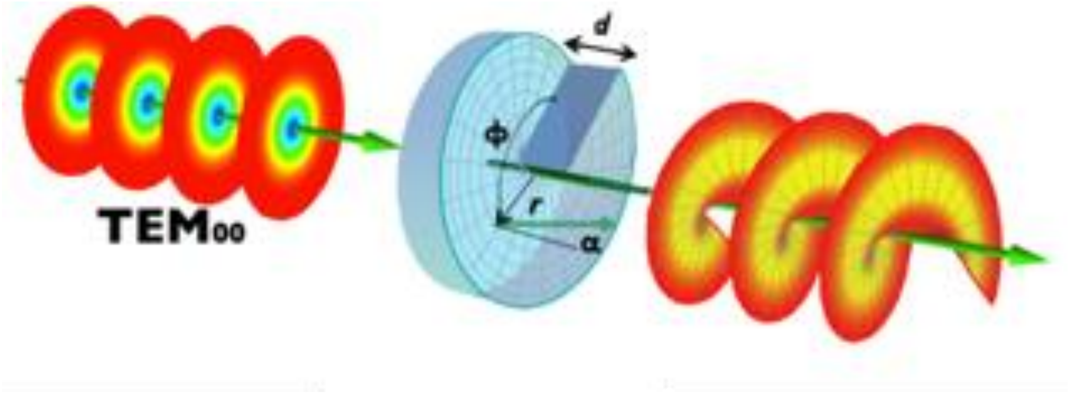
- And they used them kinetic energy spectrum of photoelectrons ejected from krypton atoms under simultaneous irradiation by soft-X-ray and visible radiation (streaking)

**Nobel Prize winner**

Prof. Ferenc Krausz

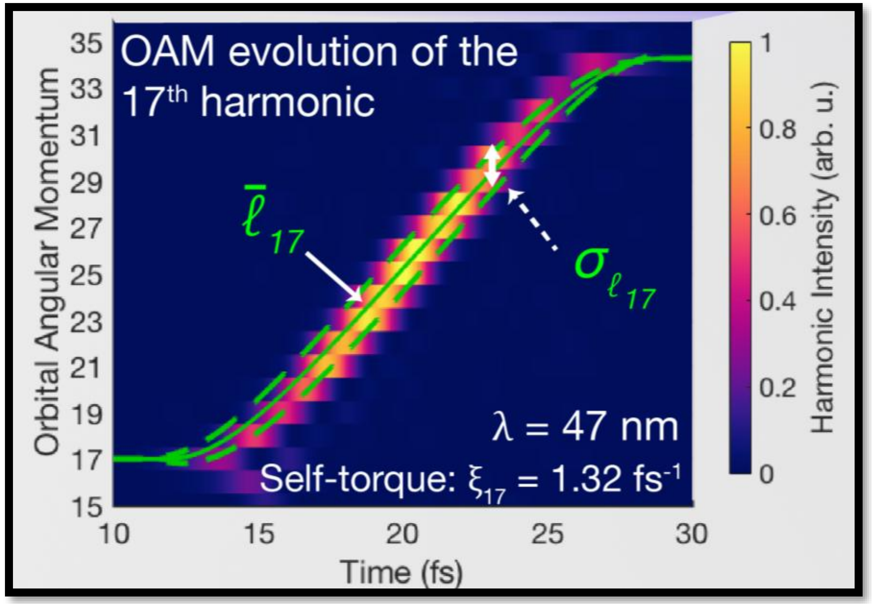
# The orbital angular momentum of light

- Vortex beams carry orbital angular momentum
- They are mathematically described by Laguerre Gauss modes
- They exhibit very promising applications
- They can be created in the optical and infrared regimes using optics elements



# Generation of light with time-dependent OAM

- The definition of the self-torque of light: $\xi_q = \frac{d\ell_q(t)}{dt}$




The time-varying orbital angular momentum:

$$\bar{\ell}_q(t) = q\{[1-\bar{\eta}(t)]\ell_1 + \bar{\eta}(t)\ell_2\}$$

$$\eta(t) = U_2(t)/U_0(t)$$

An approximation to the self-torque:

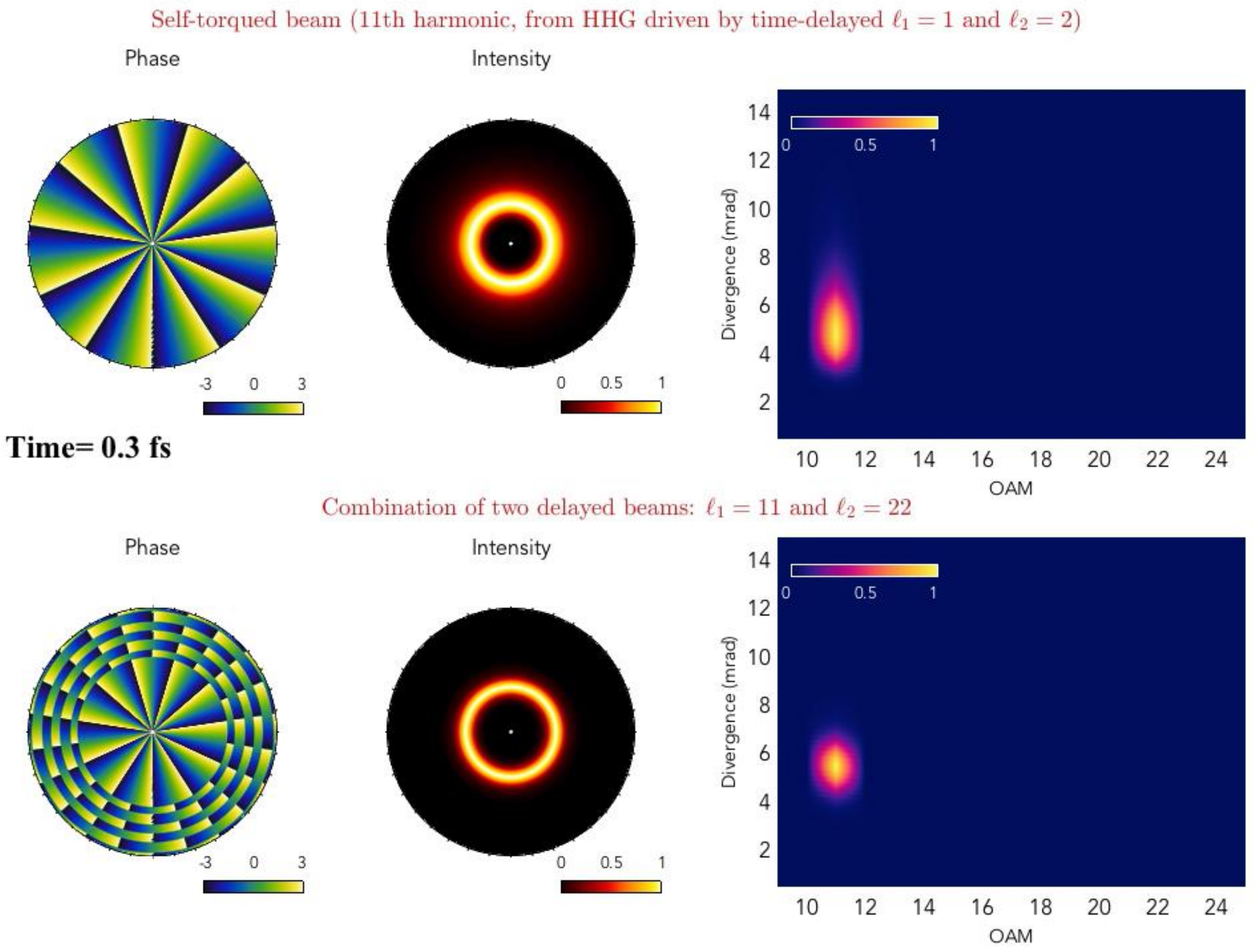
$$\hbar\xi_q = \frac{\hbar d\bar{\ell}_q(t)}{dt} = \hbar q(\ell_2-\ell_1)\frac{d\bar{\eta}(t)}{dt} \approx \hbar q(\ell_2-\ell_1)/\tau_d$$

The width of the orbital angular momentum distribution:

$$\sigma_{\ell_q} = \sqrt{\langle \ell_q^2(t)\rangle - [\langle \ell_q(t)\rangle]^2} = |\ell_2-\ell_1|\sqrt{p\bar{\eta}[1-\bar{\eta}(t)]}$$

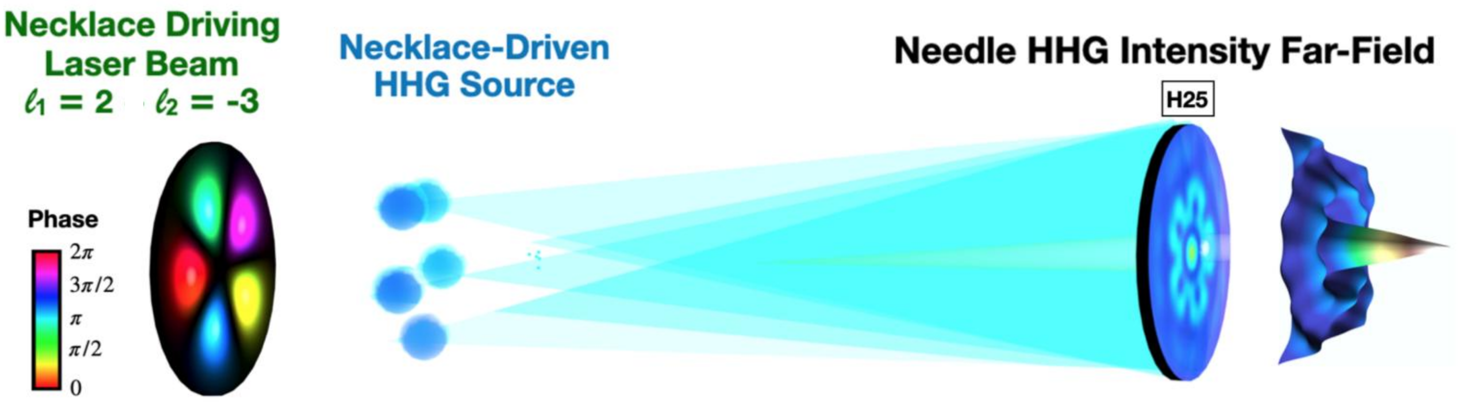
**The self-torque is well-defined!**

# Generation of light with time-dependent OAM

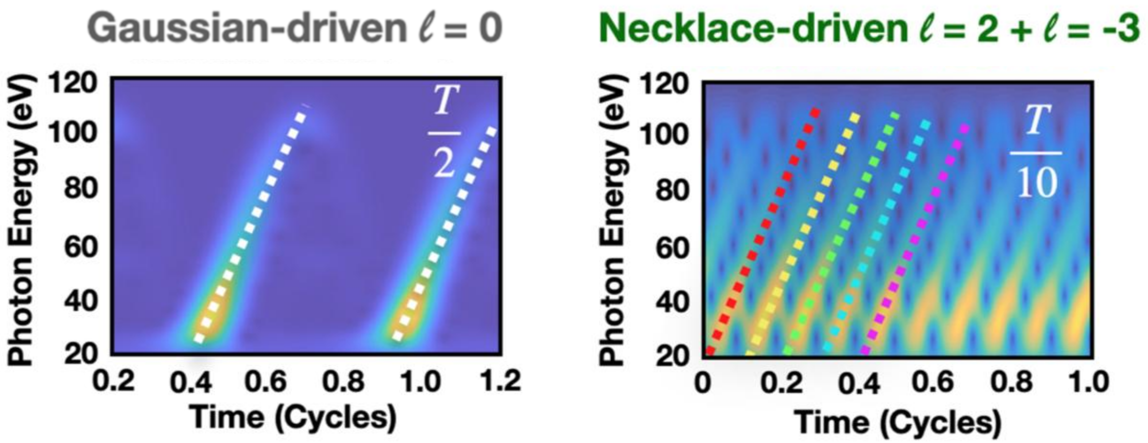


# Harmonic combs with tunable line-spacing

- ❑ HHG driven by a necklace beam: some harmonics exhibit strong on-axis yield



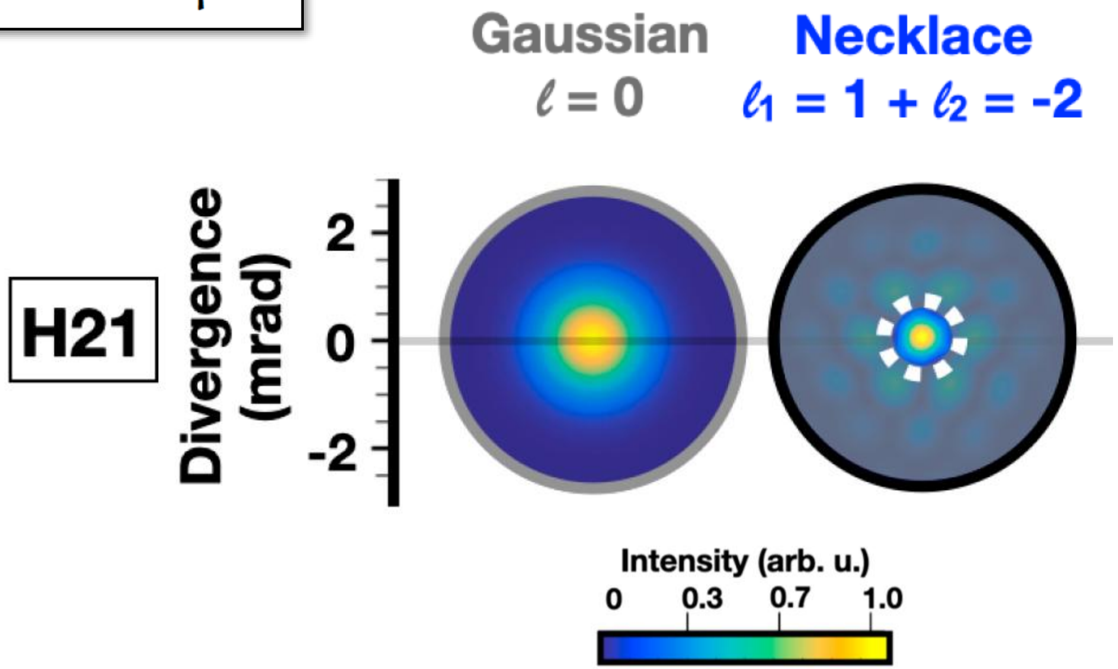

- ❑ The number of harmonic bursts per cycle detected on-axis can be manipulated



# Harmonic combs with tunable line-spacing

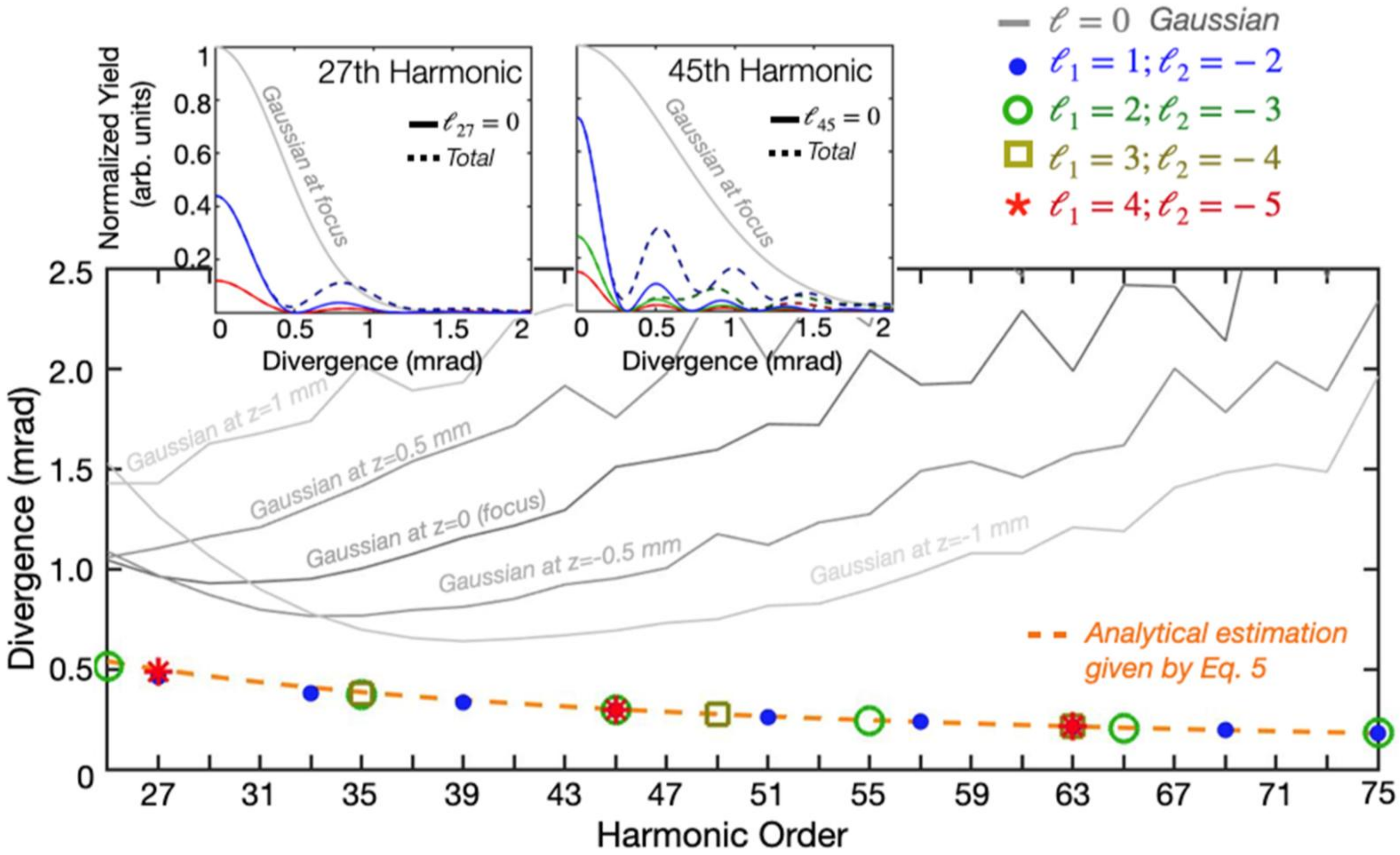
❑ The on-axis yield exhibits low divergence, which decreases with the harmonic order

$$\Delta\beta_q^{FWHM} = 2.25\frac{\lambda_0}{2\pi q R}$$



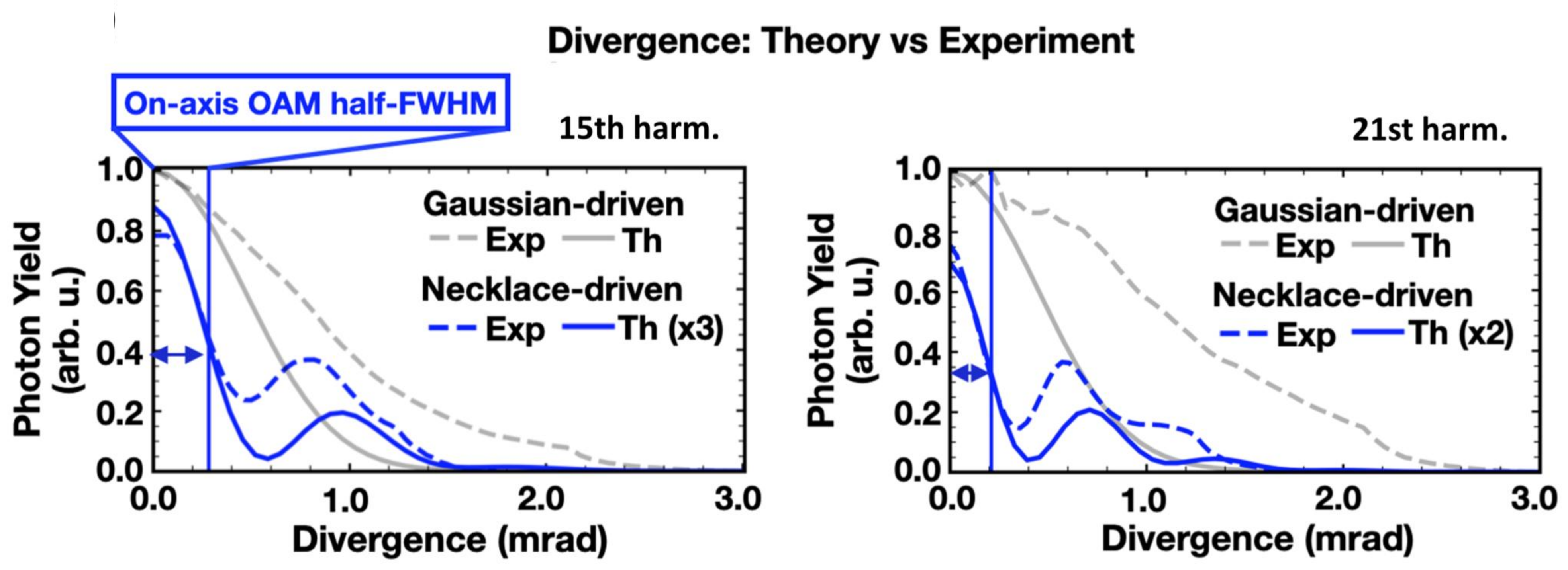
# Harmonic combs with tunable line-spacing

❑ The on-axis yield exhibits low divergence, which decreases with the harmonic order



# Harmonic combs with tunable line-spacing

- The experimental results demonstrate the generation of low-divergence frequency combs with tunable line-spacing



# HHG efficiency vs ellipticity of the driving field

❑ Different approaches for the generation of elliptically or circularly polarized harmonics:

- Anisotropic targets, such as molecules or solids

X. Zhou, et al. Physical Review Letters 102, 073902 (2009)
A. Ferré, et al. Nature Photonics 9, 93–98 (2015)
N. Tancogne-Dejean, et al. Nature Communications 8, 745 (2017)
Zurrón-Cifuentes, Ó., et al. Optics Express 27, 7776 (2019)

- Combinations of linearly polarized drivers with different frequencies

G. Lambert, et al. Nature Communications 6, 1–6 (2015)
B. Mahieu, et al. Physical Review A 97, 1–6 (2018)

- Non-collinear counter-rotating driving beams with the same color

D. D. Hickstein, et al. Nature Photonics 9, 743–750 (2015)
C. Hernandez-Garcia, et al. Physical Review A 93, 043855 (2016)
P.-C. Huang, et al, Nature Photonics 12, 349 (2018)

- Two-color, counter-rotating, circularly polarized laser fields (bi-circular driving)

H. Eichmann, et al. Physical Review A, 51, R3414–R3417 (1995)
S. Long, et al. Physical Review A, 52, 2262–2278 (1995)
D. B. Miloševic, et al. Physical Review A, 61, 063403 (2000)
A. Fleischer, et al. Nature Photonics, 8, 543–549 (2014)
O. Kfir, et al. Nature Photonics, 9, 99–105 (2015)

And more...

# Simultaneous control of orbital and spin angular momentum

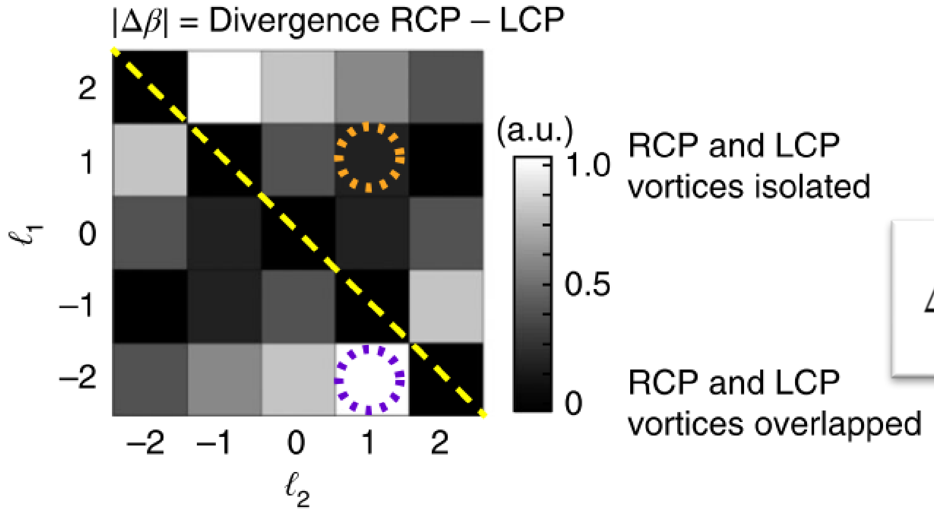
- ❑ Why are the right and left circularly polarized harmonics separated?

Selection rules

$$\begin{cases} \ell_q = n_1\ell_1 + n_2\ell_2 \\ \sigma_q = n_1\sigma_1 + n_2\sigma_2 \quad \rightarrow \quad n_2 = n_1 - \sigma_q\sigma_1 \\ q\omega_1 = n_1\omega_1 + n_2\omega_2 \end{cases}$$

$$\ell_q = \frac{q + 2\sigma_q\sigma_1}{3}(\ell_1 + \ell_2) - \sigma_q\sigma_1\ell_2$$

Divergence difference between the right and left circularly polarized harmonics

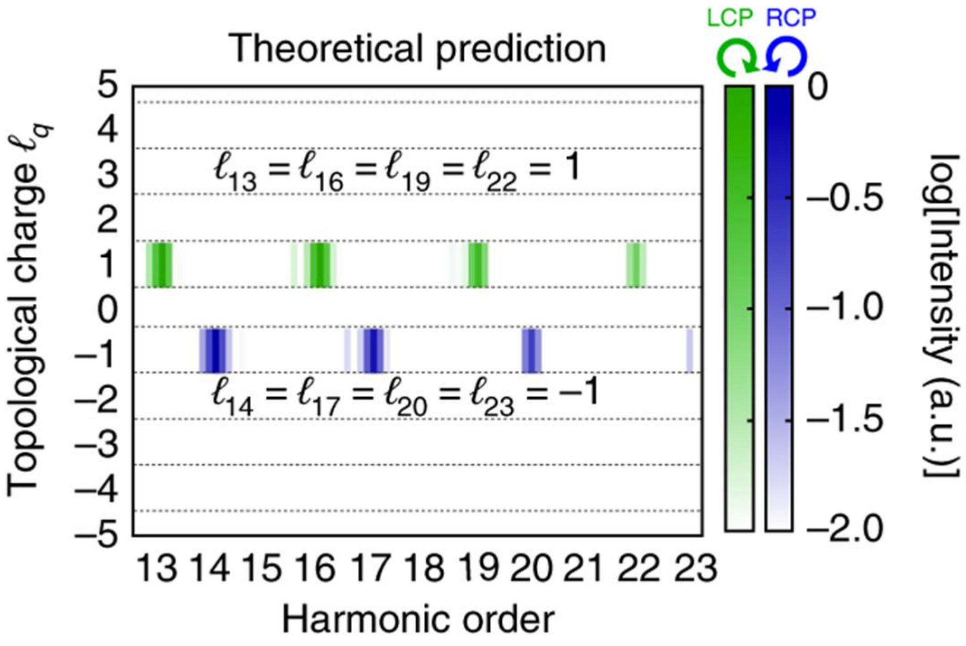


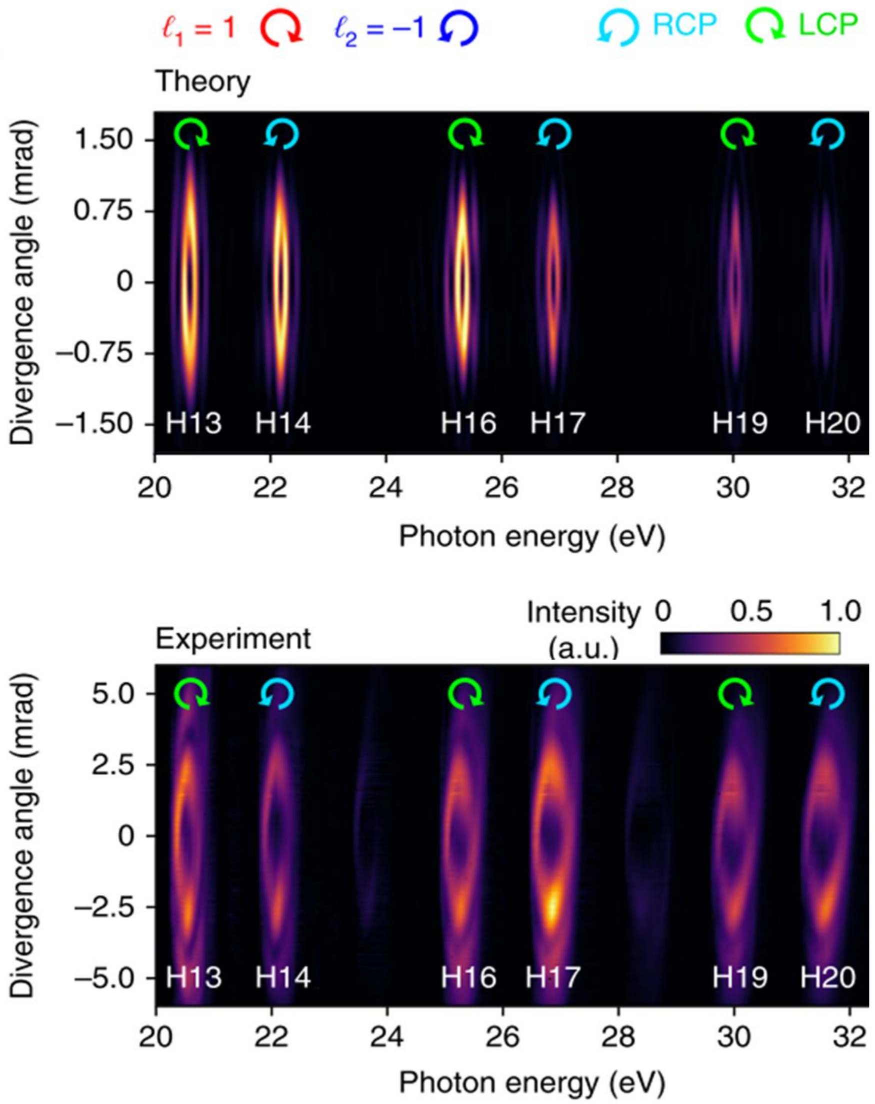

$$\Delta\beta \propto (\ell_1 - 2\ell_2)\frac{|\ell_1 + \ell_2|}{\ell_1 + \ell_2}$$

K. M. Dorney, et al. Nature Photonics, 13, 123–130 (2018)

# Simultaneous control of orbital and spin angular momentum

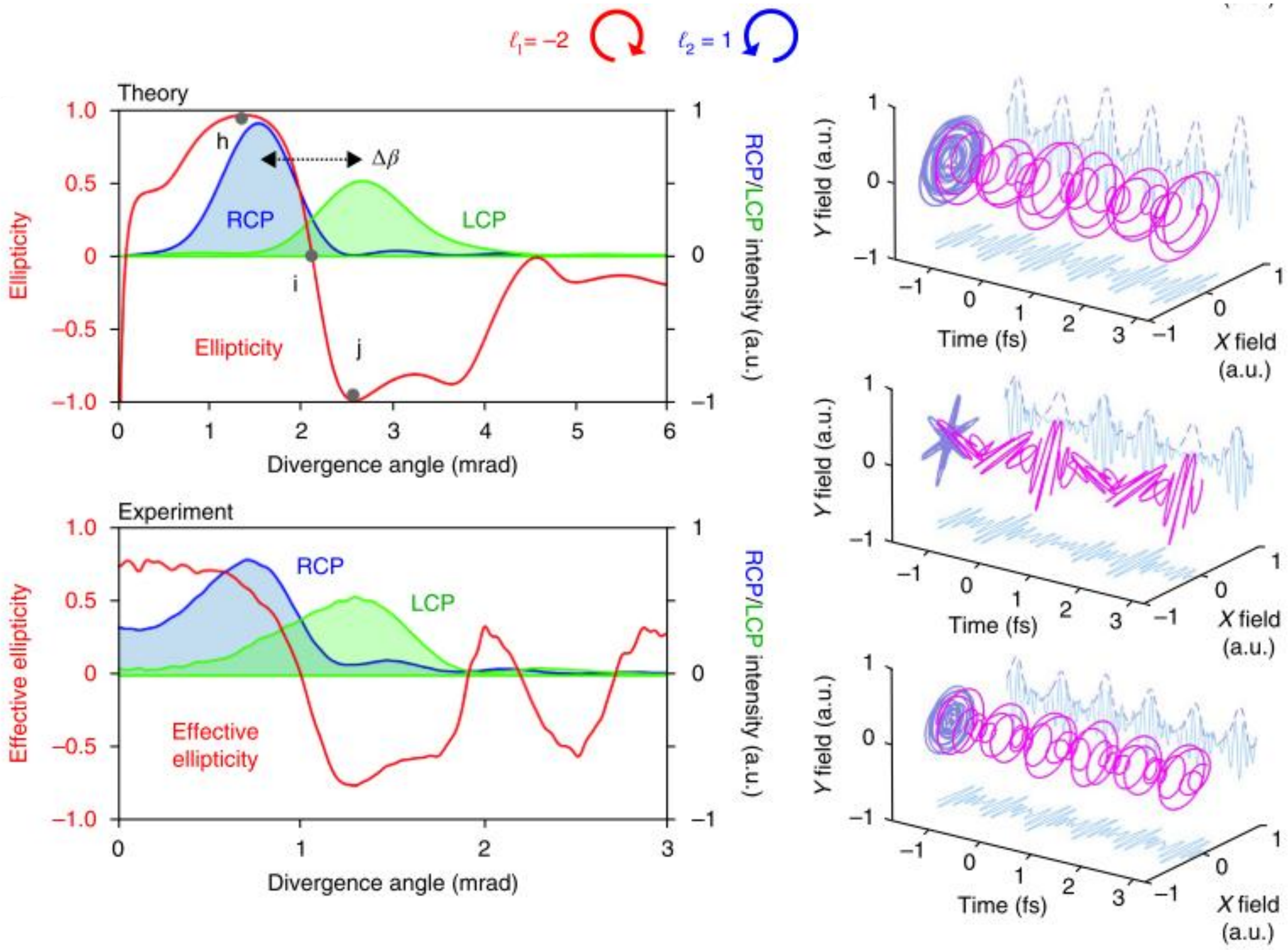
- By using driving vortex beams with opposite topological charge we generate harmonics with low orbital angular momentum



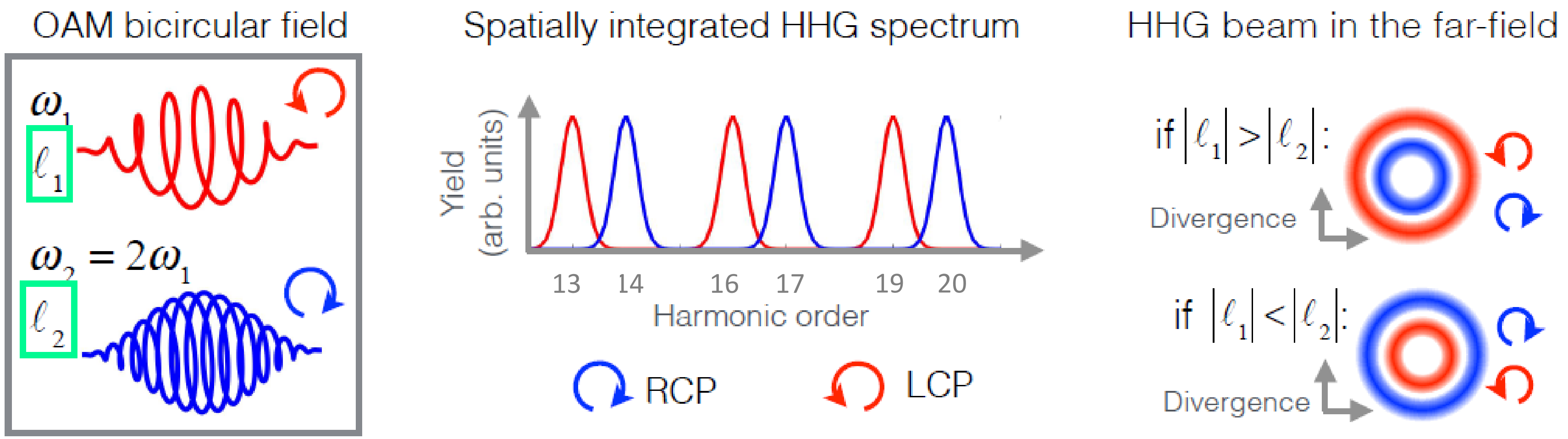
# Simultaneous control of orbital and spin angular momentum

- ❑ The polarization of the attosecond pulses is circular in the inner and outer rings

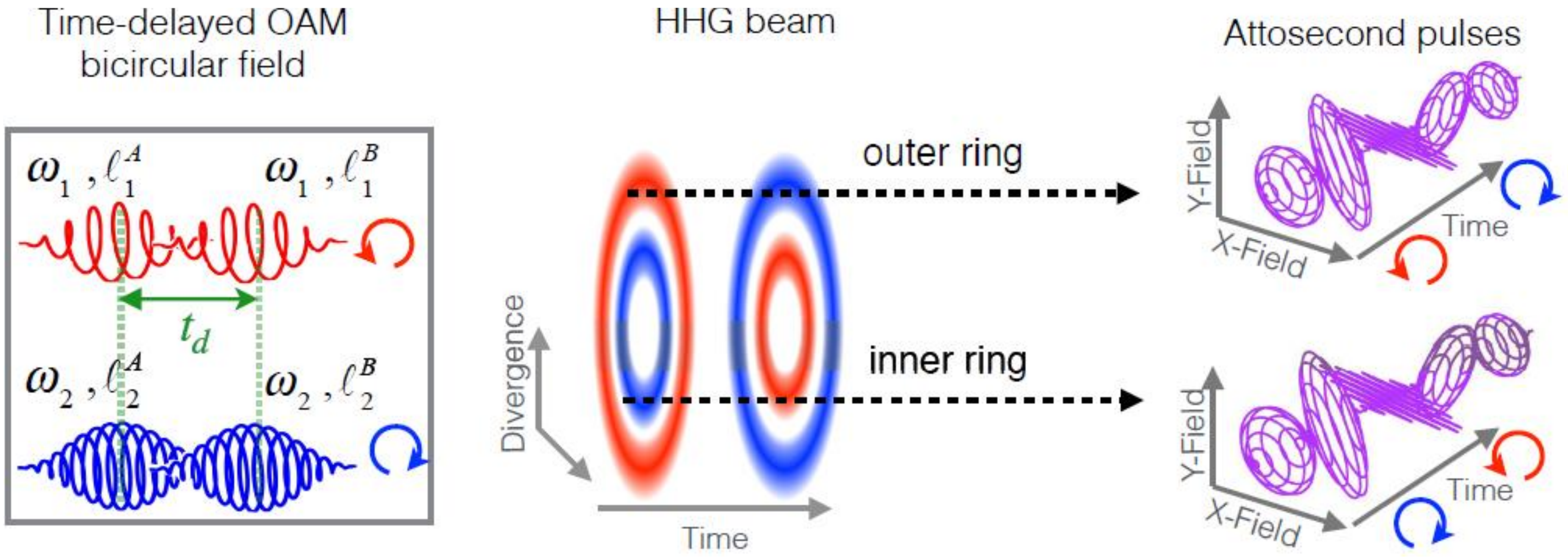


# Time-ordered polarization states in attosecond pulse trains

- The previous work:



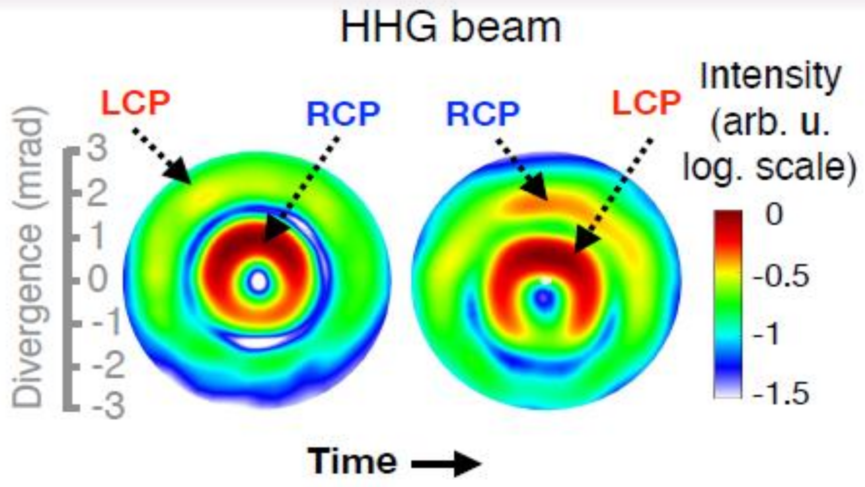

- The new scheme:



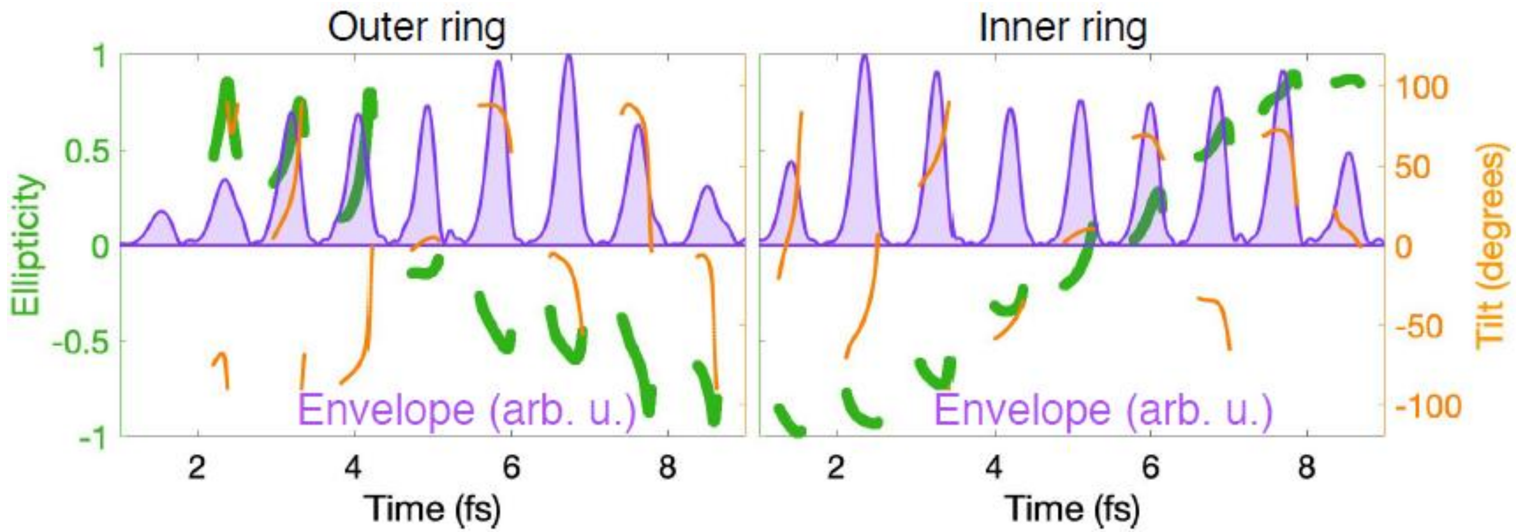
# Time-ordered polarization states in attosecond pulse trains

- ❑ Intensity profiles:

$$\ell_1^A = 2, \ell_2^A = -1, \ell_1^B = 1, \ell_2^B = -2$$




- ❑ Evolution of the ellipticity and tilt angle:

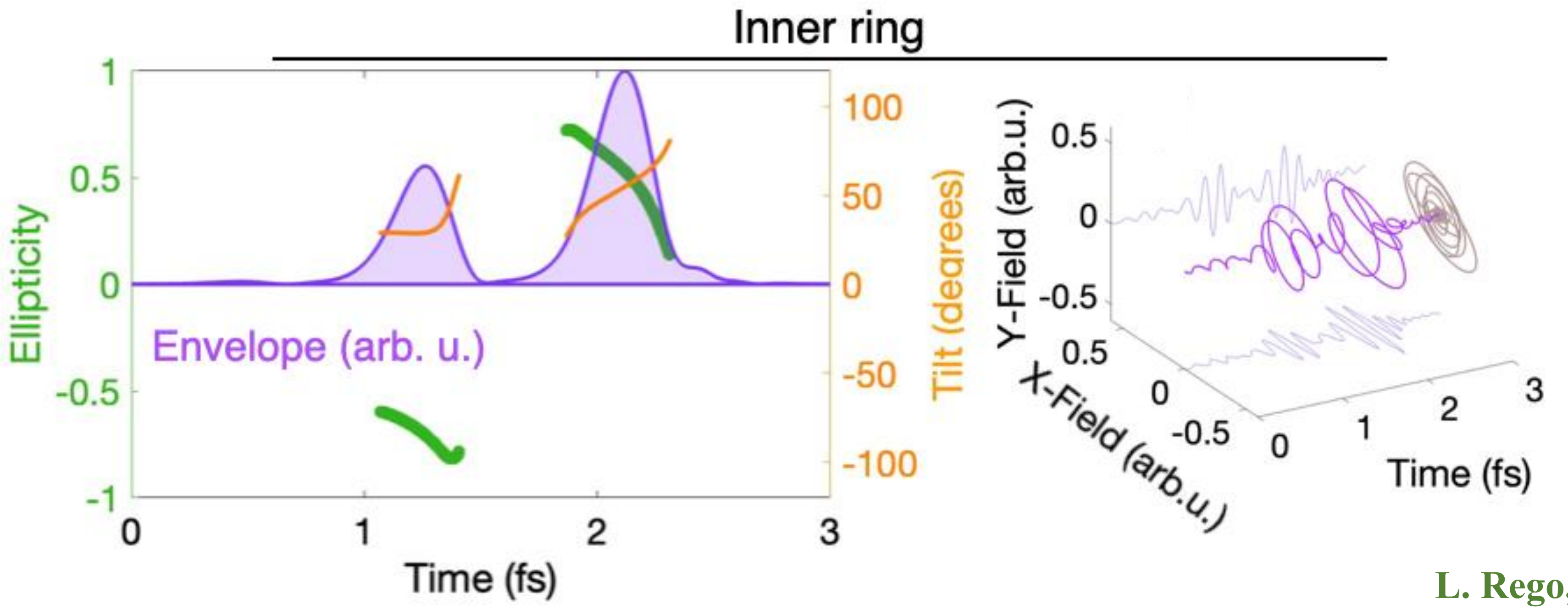



L. Rego, et al. Optics Letters, 45 (20), 5636-5639 (2020)

# Time-ordered polarization states in attosecond pulse trains

- ❑ The ultrafast evolution of the polarization state along the pulse train can be controlled via the OAM, pulse duration, and time delay of the driving fields

Using few-cycles driving pulses:




L. Rego, et al. Optics Letters, 45 (20), 5636-5639 (2020)

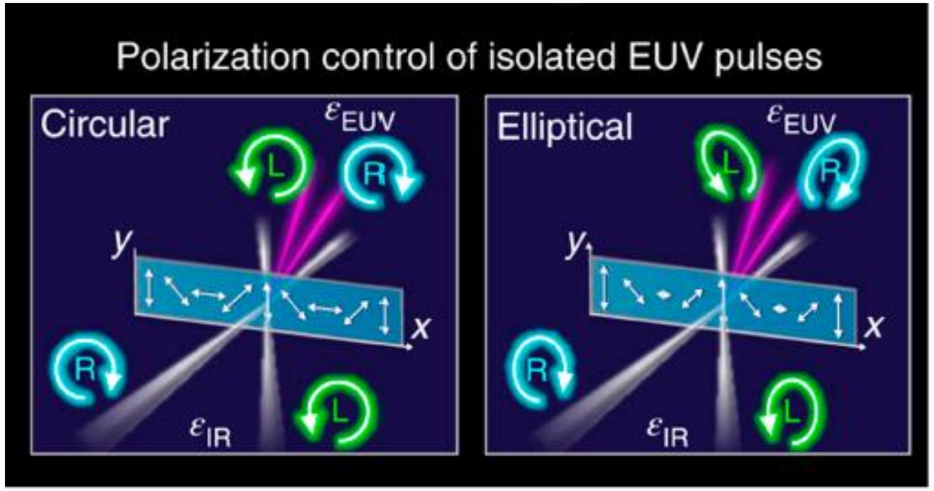
# Schemes for the creation of structured harmonic beams

## Attosecond pulses with novel polarization states

*Non-collinear counter-rotating field*

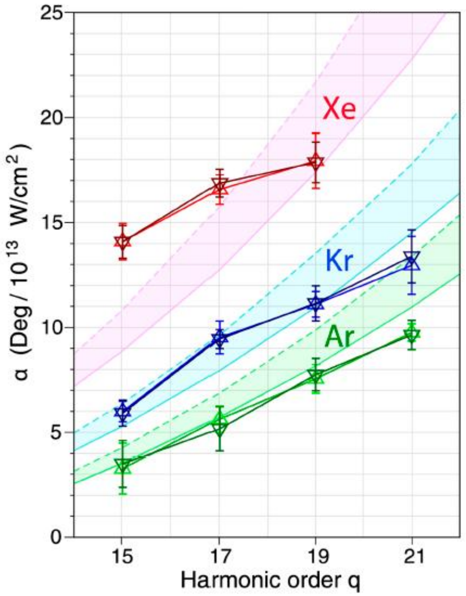
- ❑ Isolated attosecond pulses with controlled polarization

  **P.-C. Huang, et al. Nature Photonics 12, 349–354 (2018)**




- ❑ Characterization of the non-perturbative response from ellipsometry measurements

  **K.-Y. Chang, et al. Optica 8, 484 (2021)**




National Tsing Hua University (Taiwan)