Scripting the Attosecond Twist: Structured Extreme Ultraviolet Beams and Attosecond Pulses with Tailor-Made Orbital Angular Momentum

Kevin M. Dorney[1], Laura Rego[1], Nathan J. Brooks[1], Chen-Ting Liao[1], Quynh L. Nguyen[1], Julio San Román[2], Jennifer L. Ellis[1], Dmitriy Zusin[1], Christian Gentry[1], David E. Couch[1], Allison Liu[1], Emilio Pisanty[3], Justin M. Shaw[4], Antonio Picón[2], Maciej Lewenstein[3], Luis Plaja[2], Carlos Hernández-García[2], Henry C. Kapteyn[1], and Margaret M. Murnane[1]

[1]JILA - Department of Physics, University of Colorado and NIST, Boulder, Colorado, 80309, USA

[2]Grupo de Investigación en Aplicaciones del Láser y Fótonica, Departamento de Física Aplicada, Universidad de Salamanca, E-37008 Salamanca, Spain

[3]ICFO, Institut de Ciencies Fotoniques, Av. Carl Freidrich Gauss 3, 08860 Castelldefels (Barcelona), Spain

[4]Quantum Electromagnetics Division, National Institute of Standards and Technology, Boulder, Colorado 80305, USA

Abstract

• Motivation

- The angular momentum of light, present in spin and orbital flavors (SAM and OAM, respectively), can be harnessed to control light-matter interactions, enabling new technologies in super-resolution imaging, chiral sensing, and optical manipulation[1,2]. Extending these techniques to shorter wavelengths in the extreme ultraviolet (EUV) offers the potential track chiral dynamics with few-nanometer and few-fs spatiotemporal resolution.

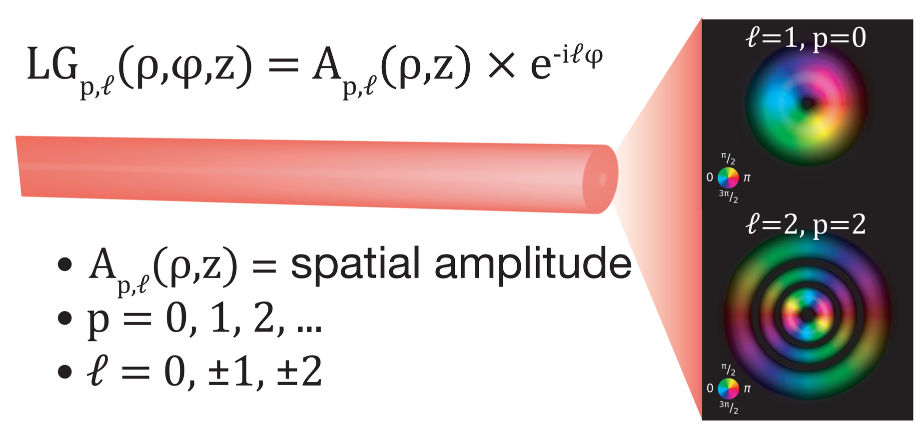
• Experiment[3,4]

- We harness the extreme quantum physics of high-harmonic generation (HHG), in order to ***synthesize EUV vortex beams —and attosecond pulses— with tailored SAM and OAM*** that rivals, and even outperforms SAM and OAM control available at optical wavelengths. By driving the HHG process with structured optical vortices in Ar gas, we realize novel EUV optical landscapes, such as ***spatially isolated EUV vortices with opposite SAM*** and ***the first demonstration of light beams possessing a time-varying OAM; the self-torque of light.***

Laguerre-Gaussian Beams: Optical Vortices

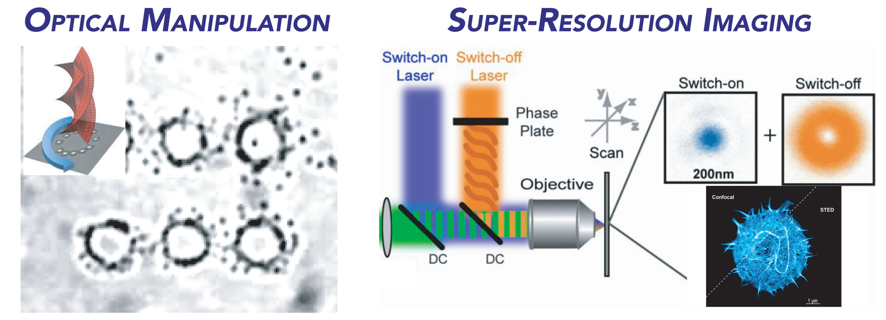
$$LG_{p,\ell}(\rho,\varphi,z) = A_{p,\ell}(\rho,z) \times e^{-i\ell\varphi}$$

- $A_{p,\ell}(\rho,z)$ = spatial amplitude
- $p = 0, 1, 2, \ldots$
- $\ell = 0, \pm1, \pm2$

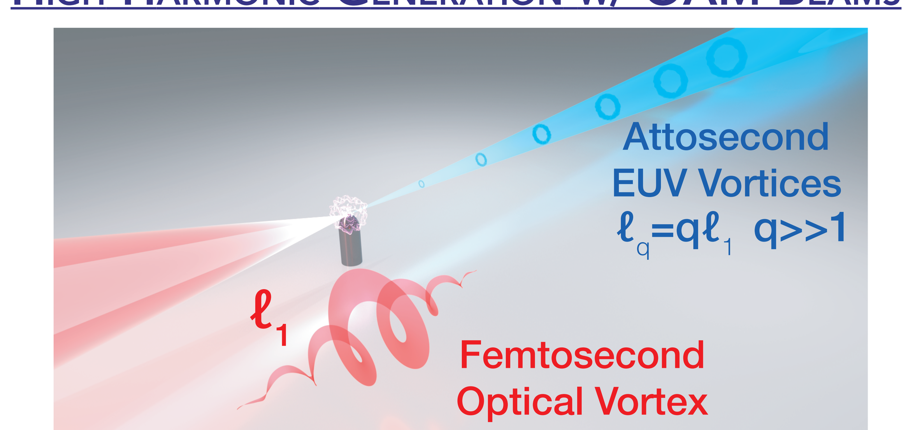


LG Beams: Versatile Optical Landscapes

Optical Manipulation — Super-Resolution Imaging



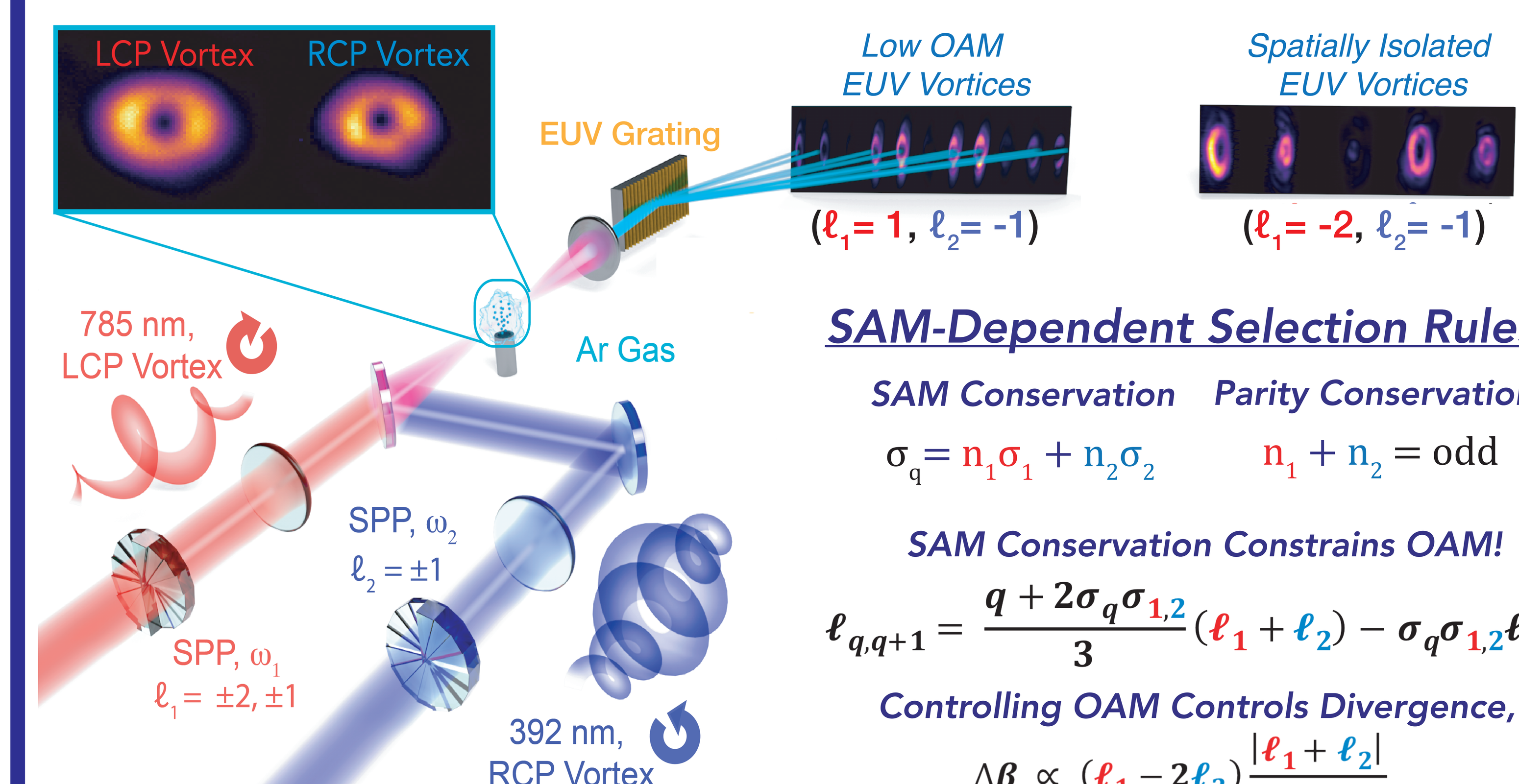
High-Harmonic Generation w/ OAM Beams



Helicity with a Twist: Attosecond Pulses with Designer SAM and OAM[3]

Summary - ***EUV beams and attosecond pulses with tailored SAM-OAM are generated by driving HHG with a "bicircular" vortex laser field*** composed of left-circularly polarized (LCP) 790 nm and right ciruclarly polarized (RCP) 395 nm vortex beams. Conservation of SAM during the HHG process restricts the amount of OAM in each harmonic, ***yielding exquisite control over the divergence, OAM charge, and polarization of the EUV vortex beams.***

High-Harmonic Generation in the Presence of SAM-OAM



SAM-Dependent Selection Rules[2,3]

SAM Conservation: $\sigma_q = n_1\sigma_1 + n_2\sigma_2$

Parity Conservation: $n_1 + n_2 = \text{odd}$

SAM Conservation Constrains OAM!

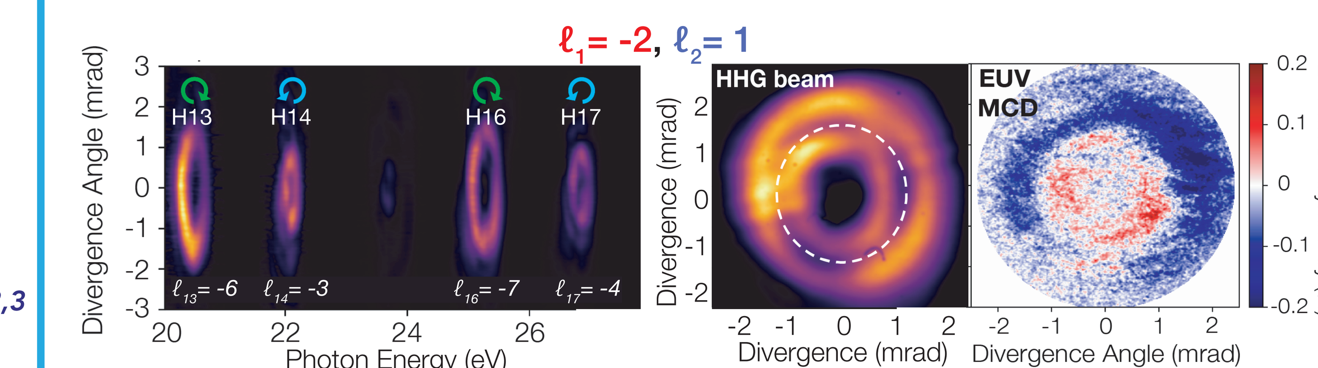
$$\ell_{q,q+1} = \frac{q + 2\sigma_q\sigma_{1,2}}{3}(\ell_1 + \ell_2) - \sigma_q\sigma_{1,2}\ell_{2,1}$$

Controlling OAM Controls Divergence, β

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

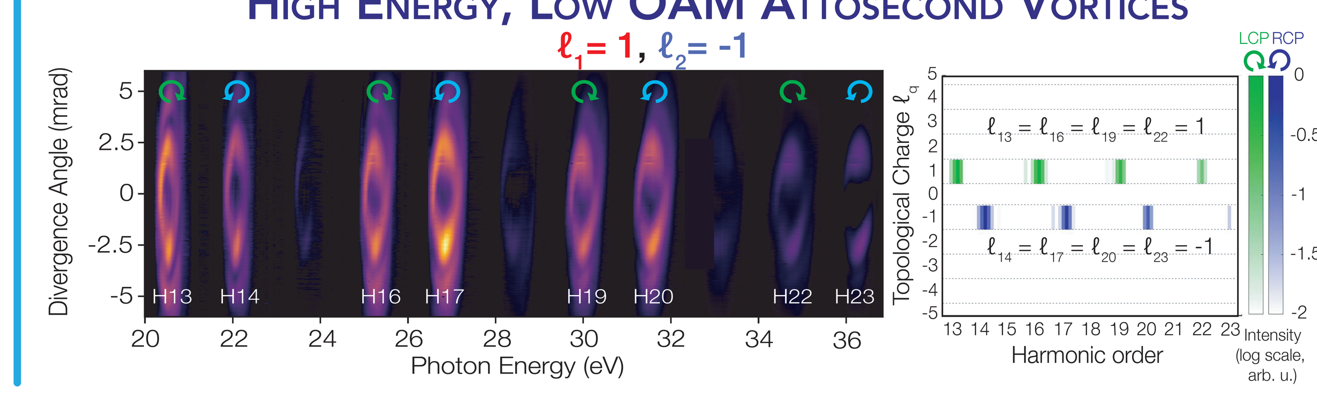
Spatially Isolated, Attosecond Vortices with Opposite Helicities

ℓ_1= -2, ℓ_2= 1



High Energy, Low OAM Attosecond Vortices

ℓ_1= 1, ℓ_2= -1

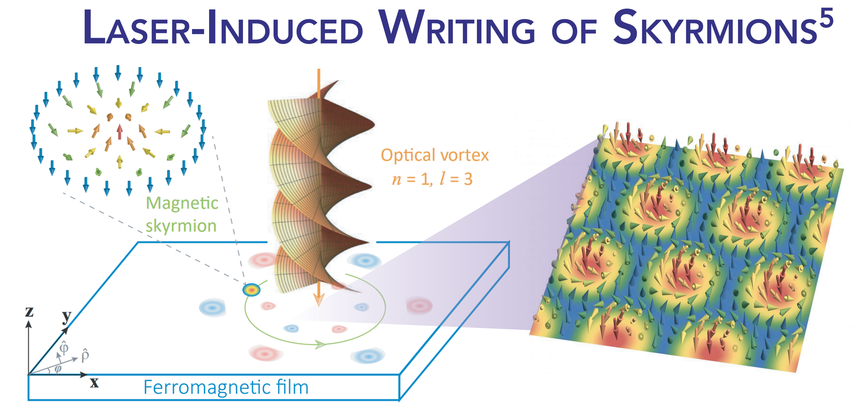


New Physics Enabled by EUV OAM Beams

• Attosecond Vortices with Tailored SAM OAM

1. First generation of EUV beams with simultaneous, controllable SAM and OAM.
2. Uniquely structured optical landscapes for ultrafast dichroism spectroscopies in the EUV.
3. Unique probe of spin-orbit interactions and topological defects on attosecond time and nm spatial scales.

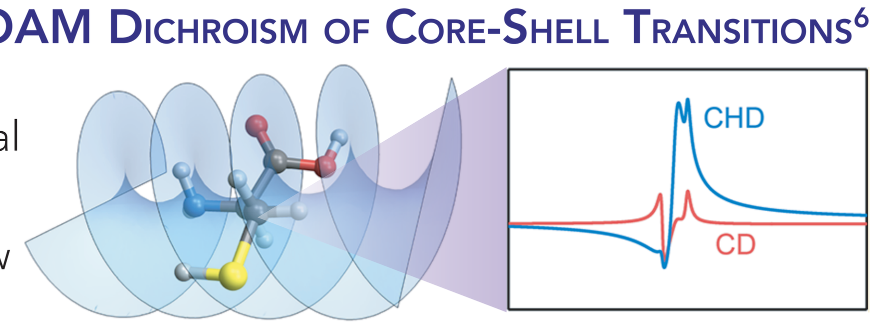
Laser-Induced Writing of Skyrmions[5]



• EUV Beams with Time-Dependent OAM

1. First prediction, observation, and control of a new optical property; self-torque, a temporal modulation of OAM.
2. Femtosecond modulation of OAM states presents a new optical tool for studying and controlling light-matter interactions.

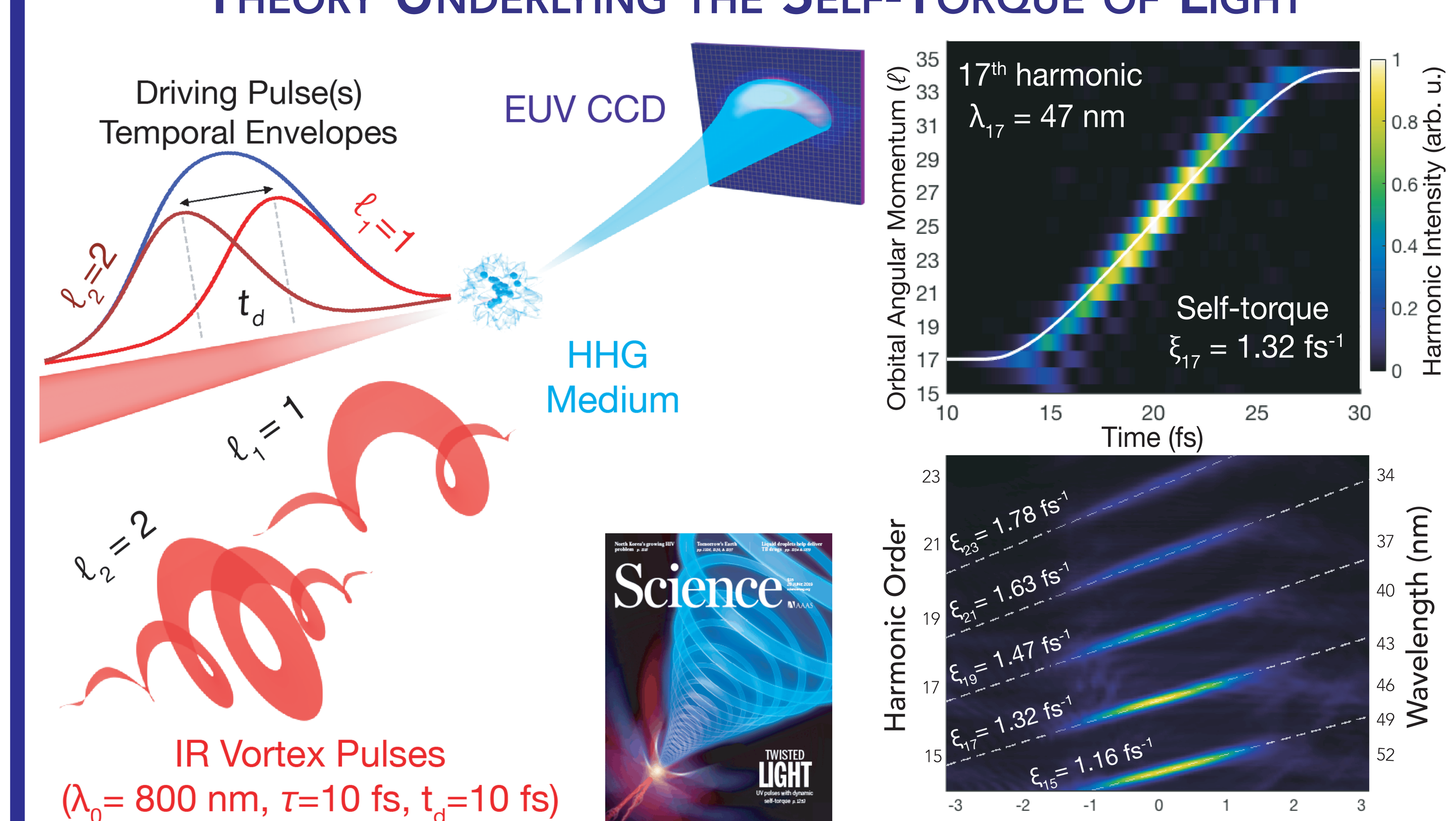
OAM Dichroism of Core-Shell Transitions[6]



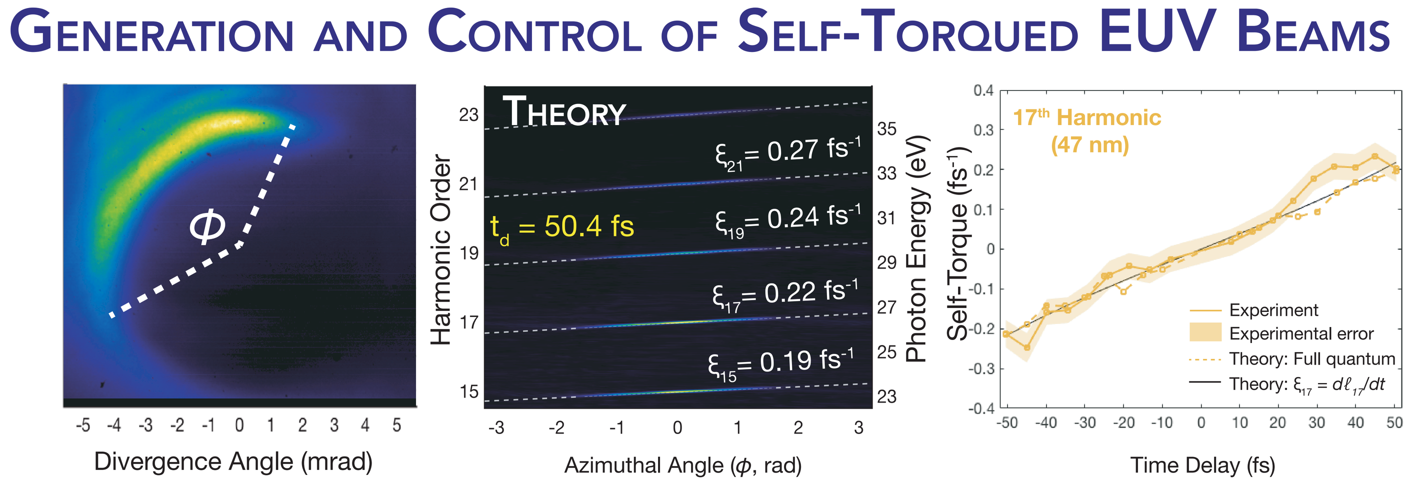
Attosecond Pulses with Time-Varying OAM: The Self-Torque of Light[4]

Summary - ***Light beams possessing self-torque —a temporal variation of their OAM— are produced and described for the first time***. By driving HHG with a pair of time-delayed infrared vortex beams with different OAM, EUV beams are emitted with an ultrafast variation of their OAM along the EUV pulse. This rapid OAM variation leads to ***unique physics in these beams, such as an azimuthal frequency chirp and time-ordered OAM states.***

Theory Underlying the Self-Torque of Light



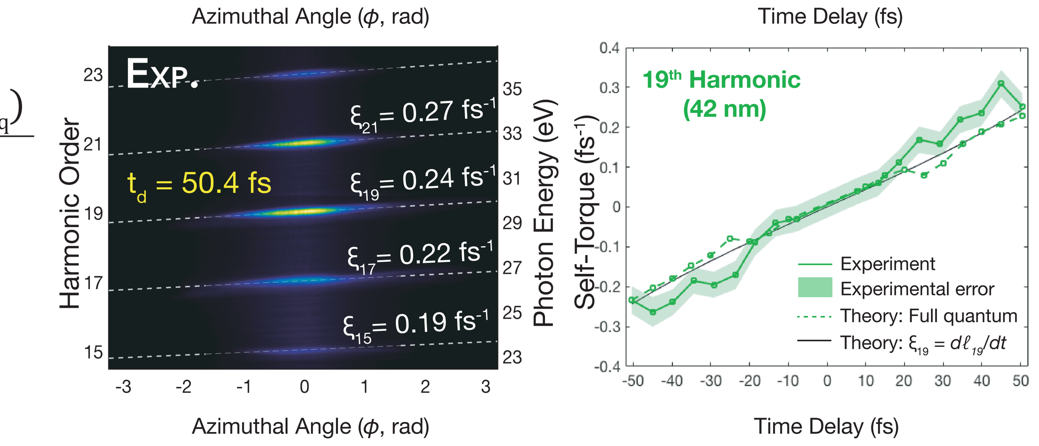
Generation and Control of Self-Torqued EUV Beams



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

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

$$\omega_q(t,\phi) \approx \omega_q + \xi_q\phi$$



Conclusions and Outlook

We have shown that ***HHG provides the ideal arena for the synthesis of EUV beams —and attosecond pulses— with unique SAM-OAM structure***. These beams, and the optical recipes that lead to their generation, can serve as ***ideal probes of ultrafast chiral dynamics in condensed-matter systems.*** The short wavelength nature of these beams makes them ***ideally suited for high-resolution imaging of topological defects at nm length scales***. At higher photon energies, SAM-OAM EUV beams could allow for spin-orbit effects to be disentangled at emental absorption edges.

References

1. Am. M. Yao, M. J. Padgett, Orbital angular momentum: origins, behavior and applications. *Adv. Opt. Photonics*, **3** (2), 2011.
2. G. Vicidomini, P. Bianchini, A. Diaspro. STED super-resolved microscopy. *Nat. Methods*, **15** (3), 2018.
3. K. M. Dorney, et al. Controlling the polarization and vortex charnge of attosecond high-harmonic beams via simultaneous spin-orbit momentum conservation. *Nat. Photonics*, **13** (2), 2019.
4. L. Rego, K. M. Dorney, et al. Generation of extreme-ultraviolet beams with time-varying orbital angular momentum. *Science* **364** (6447), 2019.
5. W. Yang, H. Yang, Y. Cao, P. Yan, Photonic orbital angular momentum transfer and magnetic skyrmion rotation. *Opt. Exp.* **26** (7), 2018.
6. L. Ye, J. R. Rouxel, S. Asban, B. Rösner, S. Mukamel. Probing molecular chirality by orbital-angular-momentum-carrying X-ray pulses. *J. Chem. Theory Comput.* **15** (7), 2019.