Spin angular momentum in extreme nonlinear optics: Controlling the polarization of high-order harmonics

Oren Cohen

Solid State Institute and Physics Department, Technion, Israel

QCN kick of meeting

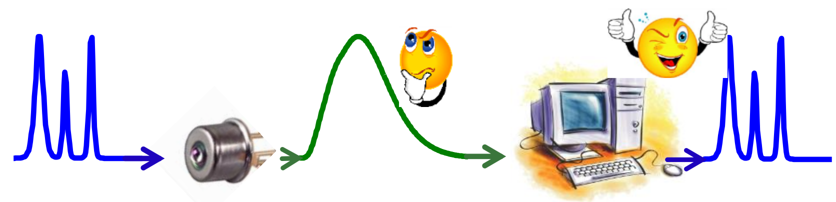
Extreme Nonlinear Optics Group @ Technion

PI: Oren Cohen

Research fellow: Dr. Avner Fleischer

PhD students: Pavel Sidorenko, Ofer Kfir & Oren Lahav

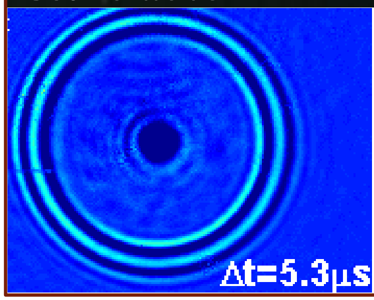
MSc students: Tzvi Diskin, Zohar Avnat & Tsachi Batkilin



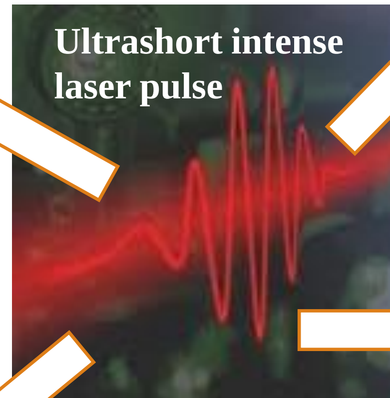
Metrology of laser pulses

Atmospheric plasma filamentation

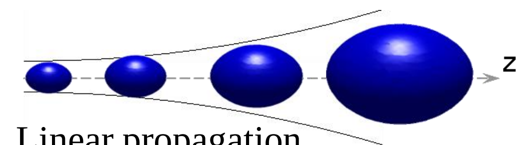
Sound wave



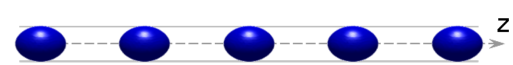
Our Vision: Induce photonic structures in air from remote



Spatio-temporal solitons: Light bullets



Linear propagation

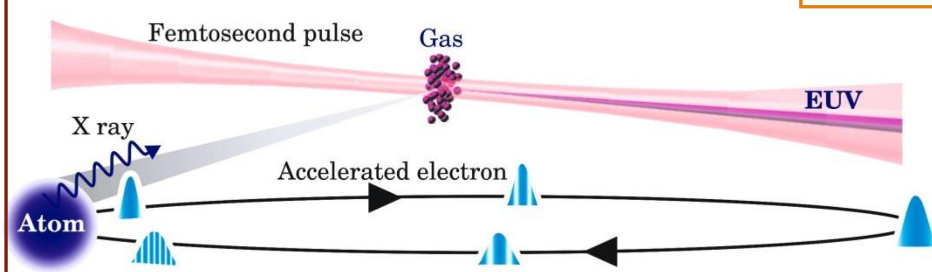


Nonlinear propagation: 3D solitons

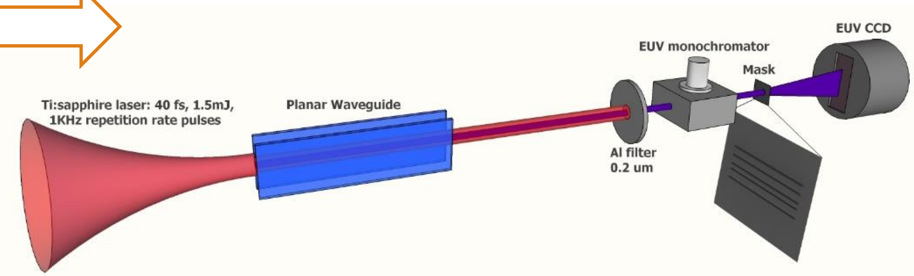
Frequency up-conversion to extreme UV and X-rays

Vision: - enhance the process efficiency

- control the EUV & X-rays properties



Develop ultrafast nano-imaging techniques & applications



Spin angular momentum in extreme nonlinear optics

Avner Fleischer, Ofer Kfir, Tzvi Diskin, Pavel Sidorenko and Oren Cohen

Energy conservation

$$\Omega_{HHG} = n_1 \cdot \omega_1 + n_2 \cdot \omega_2$$

$$\Updownarrow$$

$$\sigma_{HHG} = n_1 \cdot \sigma_1 + n_2 \cdot \sigma_2$$

Spin conservation $|\sigma| \leq 1$

<u>Experiment</u>: mixing of waves with controlled polarizations

Spin angular momentum in extreme nonlinear optics

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<u>Experiment</u>: mixing of waves with controlled polarizations

<u>Main achievements</u>

- **Spin angular momentum in XNLO**
- **Full control over polarization of high-order harmonics using a simple knob, without compromising efficiency.**
- **Missing quanta for conservation of angular momentum**

Spin angular momentum in extreme nonlinear optics

Avner Fleischer, Ofer Kfir, Tzvi Diskin, Pavel Sidorenko and Oren Cohen

Energy conservation

$$\Omega_{HHG} = n_1 \cdot \omega_1 + n_2 \cdot \omega_2$$

$$\Updownarrow$$

$$\sigma_{HHG} = n_1 \cdot \sigma_1 + n_2 \cdot \sigma_2 + \delta$$

Spin conservation $|\sigma| \leq 1$

<u>Experiment</u>: mixing of waves with controlled polarizations

<u>Main achievements</u>

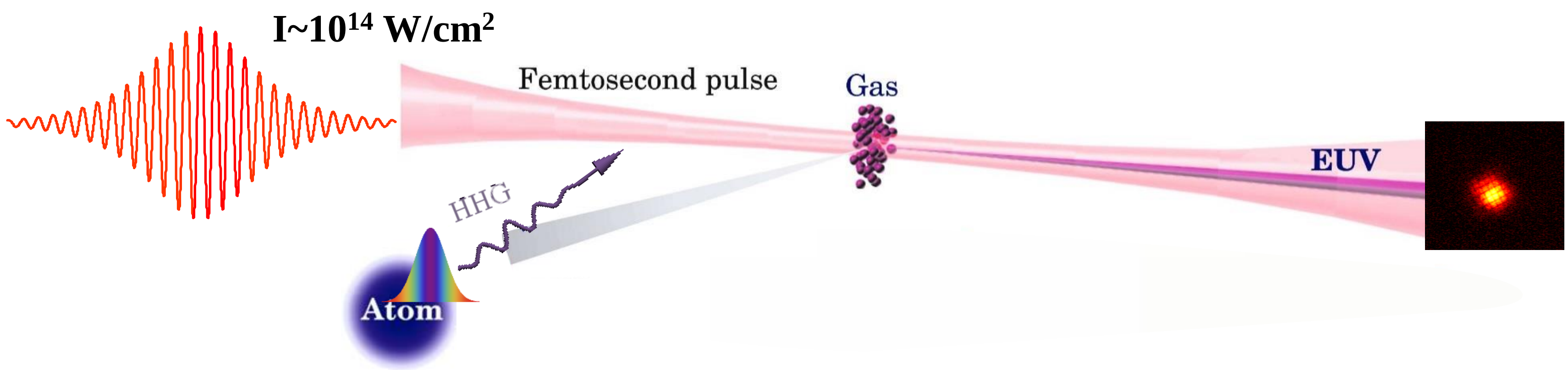
- **Spin angular momentum in XNLO**
- **Full control over polarization of high-order harmonics using a simple knob, without compromising efficiency.**
- **Missing component for conservation of angular momentum**

Outline

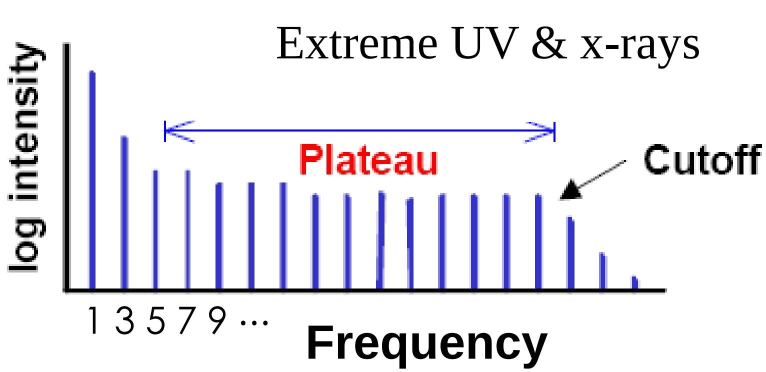
- Introduction to extreme nonlinear optics
- High harmonic generation:
  - Polarization
- Controlling the polarization of HHG
- Spin angular momentum conservation

High Harmonic Generation

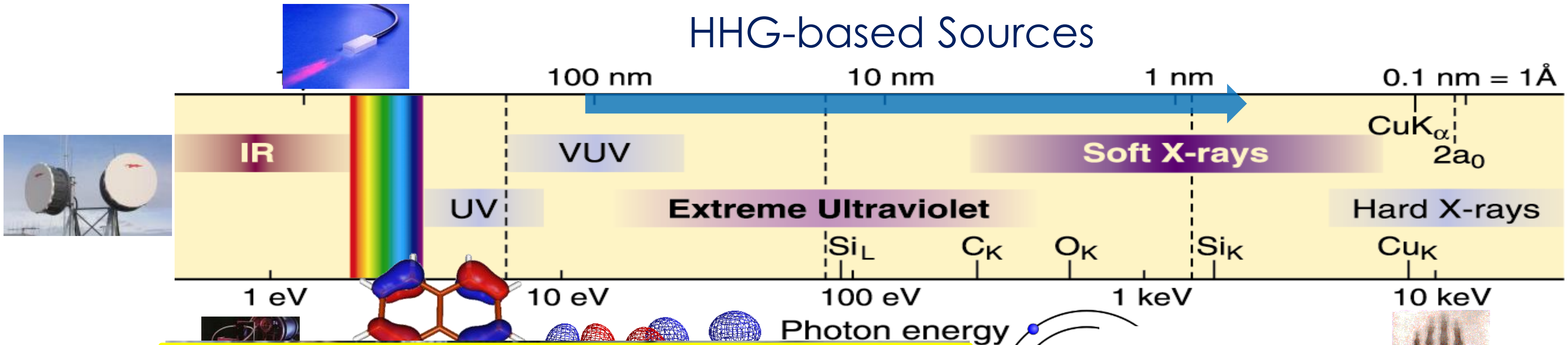
- Intense femtosecond pulse interacts with a gas generates high harmonics.

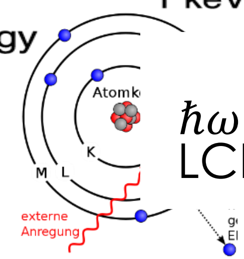


A. McPherson *et al.*, JOSA B 4, 595 (1987)
M. Ferray et al., J. of Phys. 21, L31 (1988).
Kulander, K. C., *et al*. Laser Physics **3**, 359 (1993)
P. B. Corkum, PRL **71**, 1994 (1993)
M. Lewenstein et al., PRA 49, 2117 (1994)



Sources of Extreme UV & X-rays

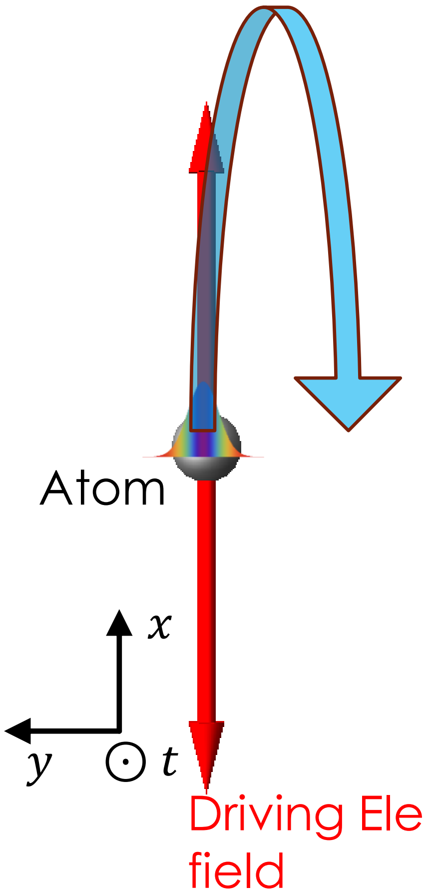




$\hbar\omega$~10 keV
LCLS-SLAC

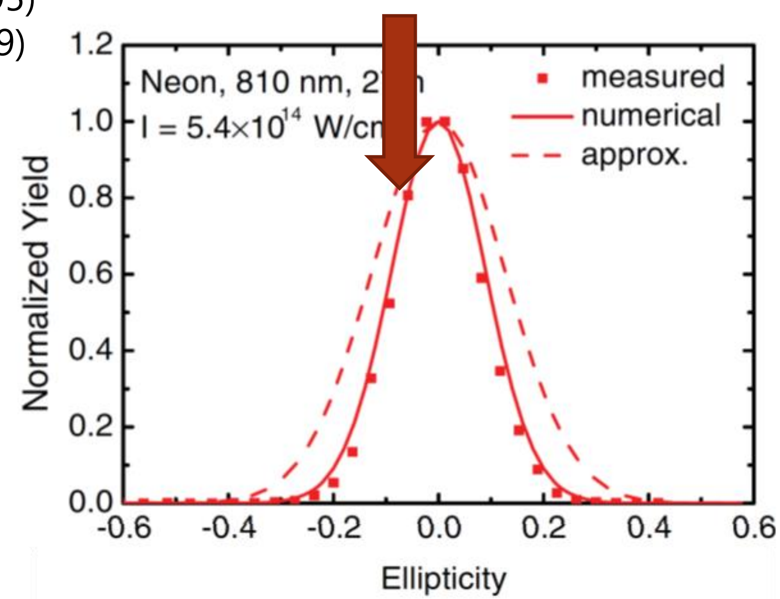
BESSY
Synchrotron

Ellipticity Effect



- High harmonic radiation commonly composed of **odd harmonics**, with **linear polarization** $\varepsilon \approx 0$.
- Ellipticity diminishes HHG efficiency
- maximal measured HHG ellipticity $\varepsilon < 0.4$

Weihe, F.A., *et al.*, PRA **51**, R3433 (1995)
Zhou, X., *et al.*, PRL **102**, 073902 (2009)



Möller, M., *et al.* PRA **86**, 011401 (2012)

Circularly & Elliptically Polarized HHG

Circularly polarized HHG

Long,

Miloš

Alon,

Nobu

Yuan,

Lewis

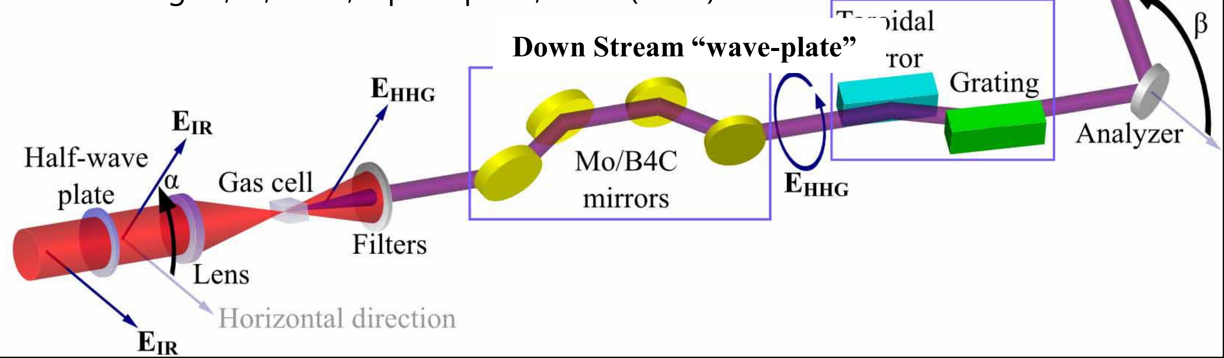
Husa... Opt. Exp. ...

Yuan & Bandrauk, PRL 110 023003 (2013)

Using an extreme-UV "wave-plate"

Transmission<4%

Vodungbo, B., *et al.*, Opt. Exp. **19**, 4346 (2011)



Elliptically polarized HHG

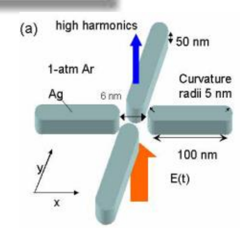
Weihe, F. A., *et al.*, PRA **51**, R3433 (1995)

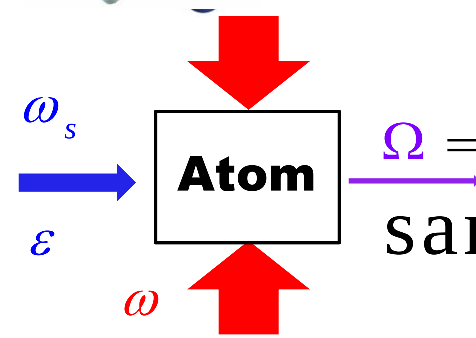
Strelkov, V. V. *et al.*, PRL **107**, 043902 (2011)

Zhou, X., *et al.*, PRL **102**, 073902 (2009)

Yuan, K. J. and Bandrauk., A. D.,PRA **83**, 063422 (2011)

Fleischer, A., *et al.*, OL **38**, 223 (2013)





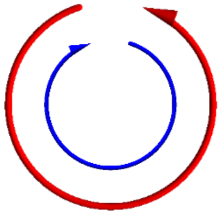
$k \in \mathbf{Z}$

$\Omega = \pm\omega_s + 2k\omega$

same ε

Circularly & Elliptically Polarized HHG

Circularly polarized HHG



Long, S., *et al.*, PRA **52**, 2262 (1995)
Pioneering experiment by Eichmann, H. *et al.*, PRA **51**, R3414 (1995)

Milošević, D. B., *et al.*, PRA **61**, 063403 (2000)

Alon, O., *et al.*, PRL **80**, 3743 (1998)

Nobusada, K., and Yabana, K., PRA **75**, 032518 (2007)

Yuan, K. J., *et al.*, PRA **84**, 023410 (2011)

Lewis, Z. L., *et al.*, OL **37**, 2415 (2012)

Husakou, A., Opt. Exp. **19**, 25346 (2011)

Yuan & Bandrauk, PRL 110 023003 (2013)

Elliptically polarized HHG

Weihe, F. A., *et al.*, PRA **51**, R3433 (1995)

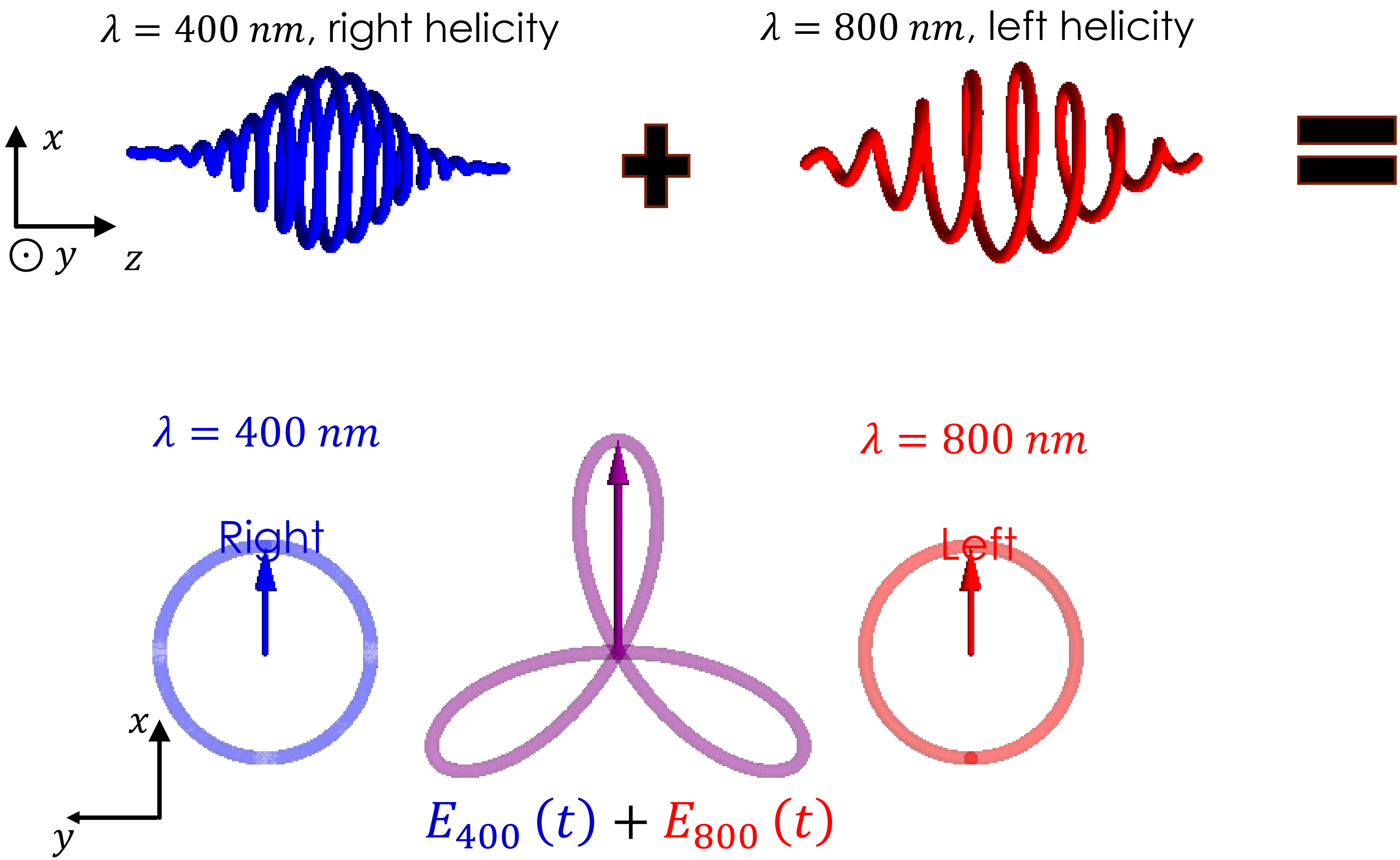
Strelkov, V. V. *et al.*, PRL **107**, 043902 (2011)

Zhou, X., *et al.*, PRL **102**, 073902 (2009)

Yuan, K. J. and Bandrauk., A. D.,PRA **83**, 063422 (2011)

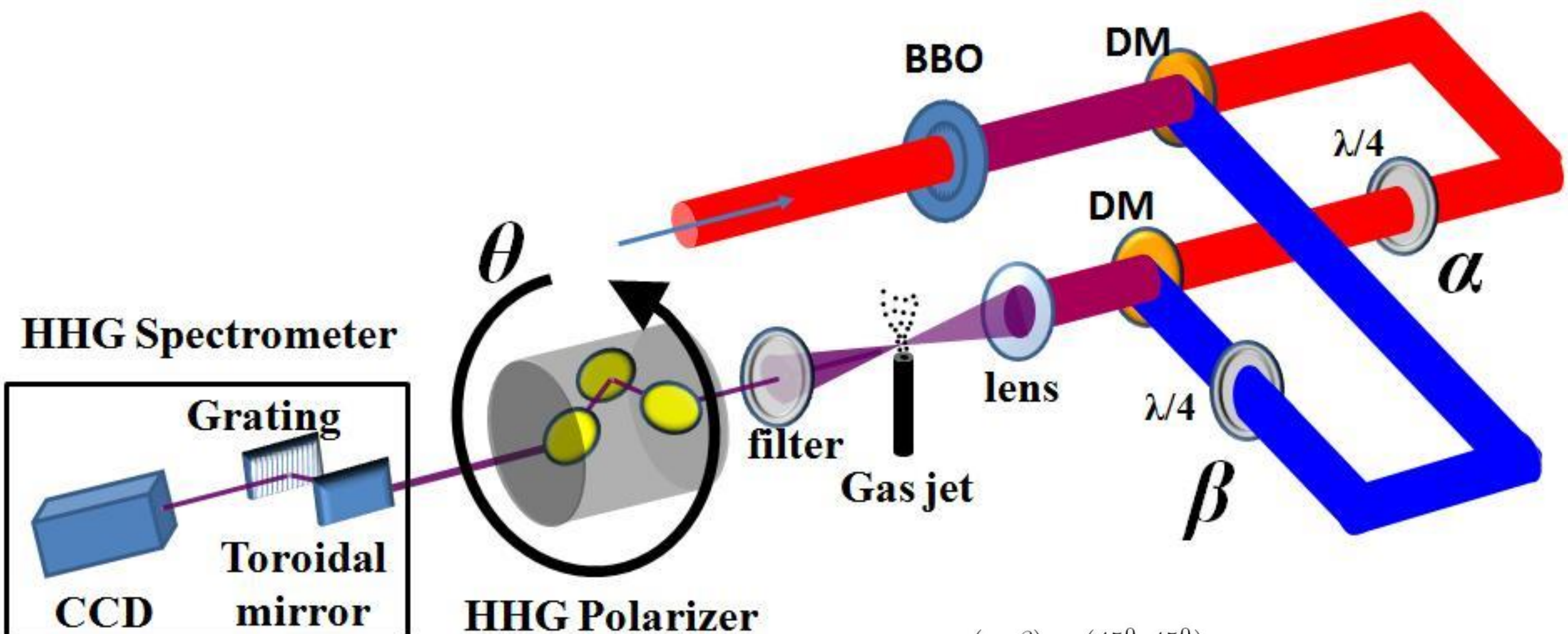
Fleischer, A., *et al.*, OL **38**, 223 (2013)

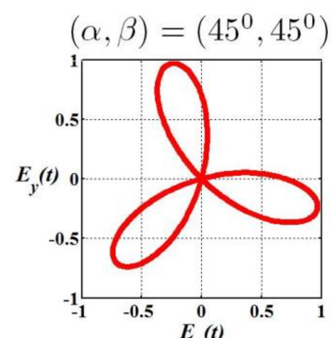
Counter-Rotating Bi-Chromatic Driver

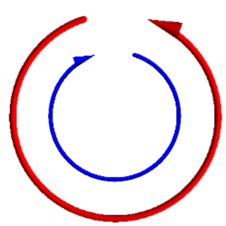


Long, S. *et al.*, PRA **52** 2262 (1995).

High harmonic generation with counter-rotating circularly-polarized bi-chromatic fields

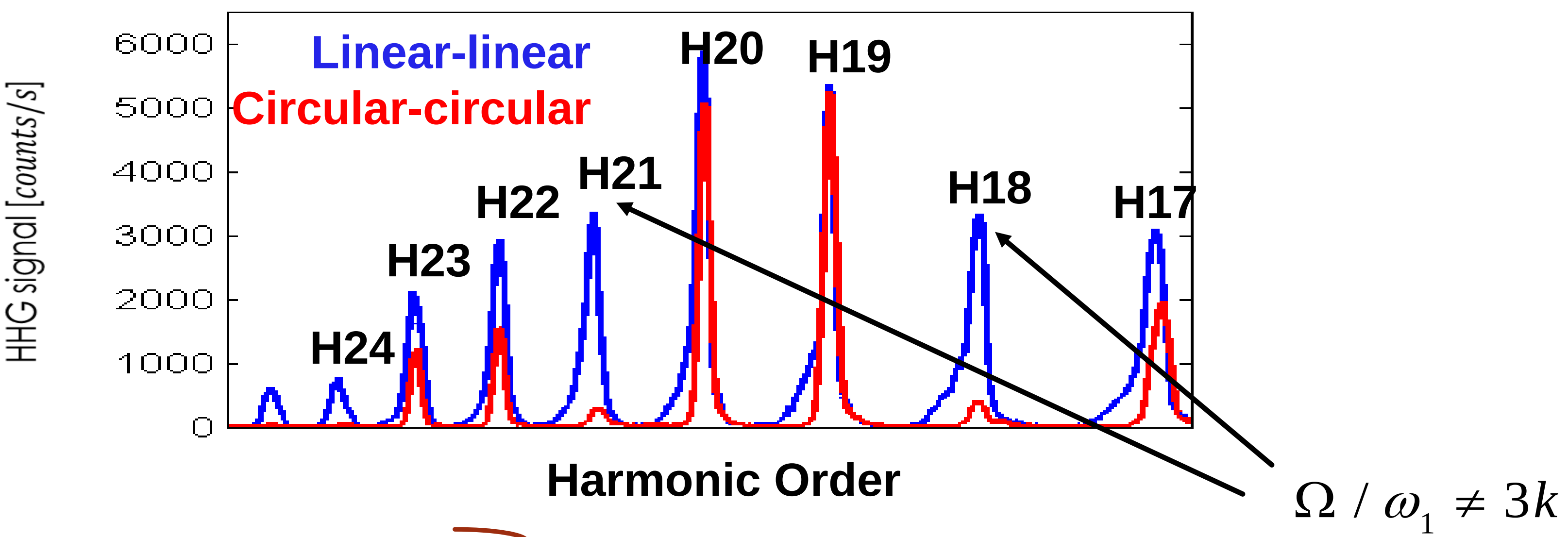






Comparable efficiency to HHG by lineally polarized driver

HH signal for 1.2W red & 0.67W blue



$$\Omega_{HHG} = n_1 \cdot \omega_1 + n_2 \cdot 2\omega_1$$

$$\sigma_{HHG} = n_1 \cdot \sigma_1 + n_2 \cdot \sigma_2$$

Spin conservation $|\sigma| \leq 1$

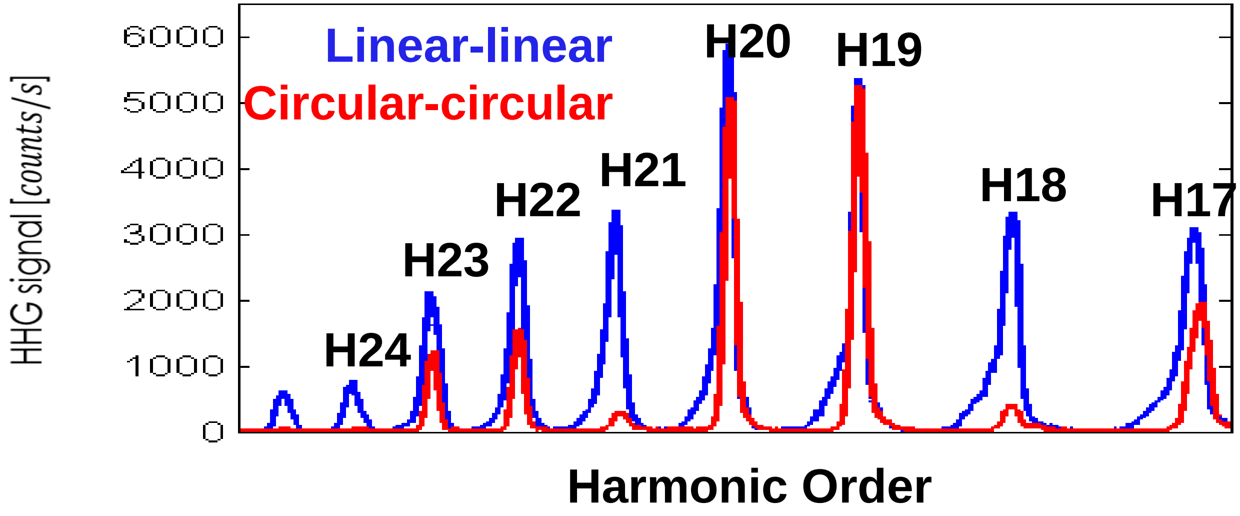
$n_1 + n_2 =$ odd integer

No solutions with

$$\Omega_{HHG} / \omega_1 = 3k$$

Comparable efficiency to HHG by lineally polarized driver

HH signal for 1.2W red & 0.67W blue



$$\Omega_{19H} = 7\omega_1 + 6 \cdot 2\omega_1$$

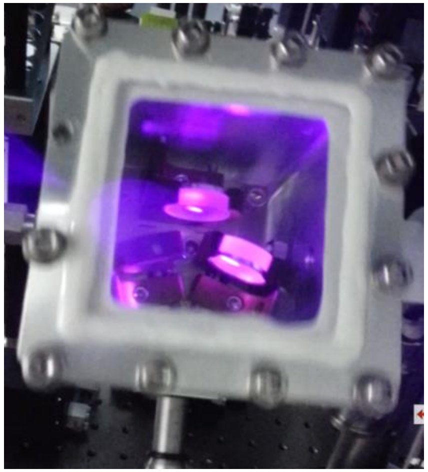
$$\sigma_{19H} = 7 \cdot (1) - 6 \cdot (-1) = 1$$

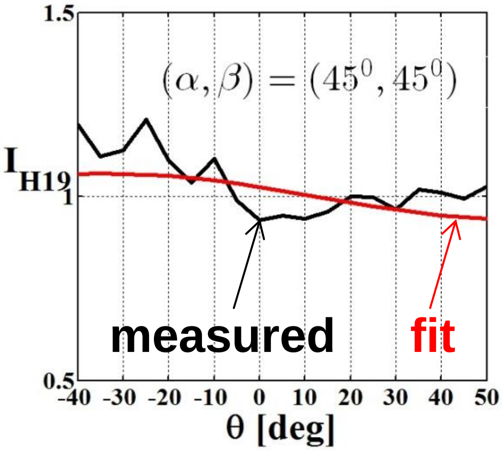
$$\Omega_{20H} = 6\omega_1 + 7 \cdot 2\omega_1$$

$$\sigma_{20H} = 6 \cdot (1) - 7 \cdot (-1) = -1$$

Circularly-polarized HHG

- **Measured circularity**





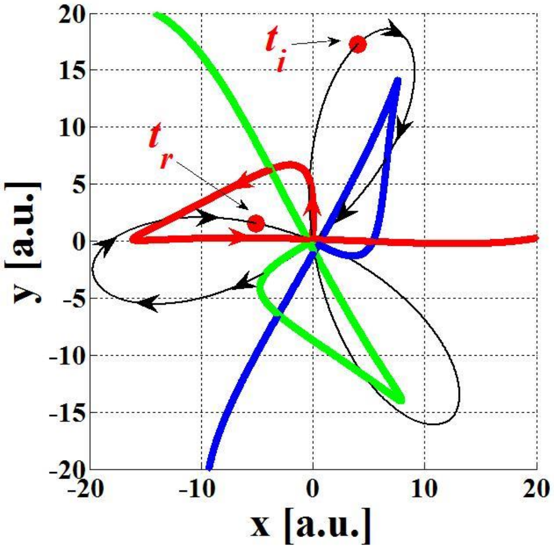
$$\Rightarrow \quad \varepsilon_{19} = 0.95$$

$$\varepsilon_{19}^{theo} = 1$$

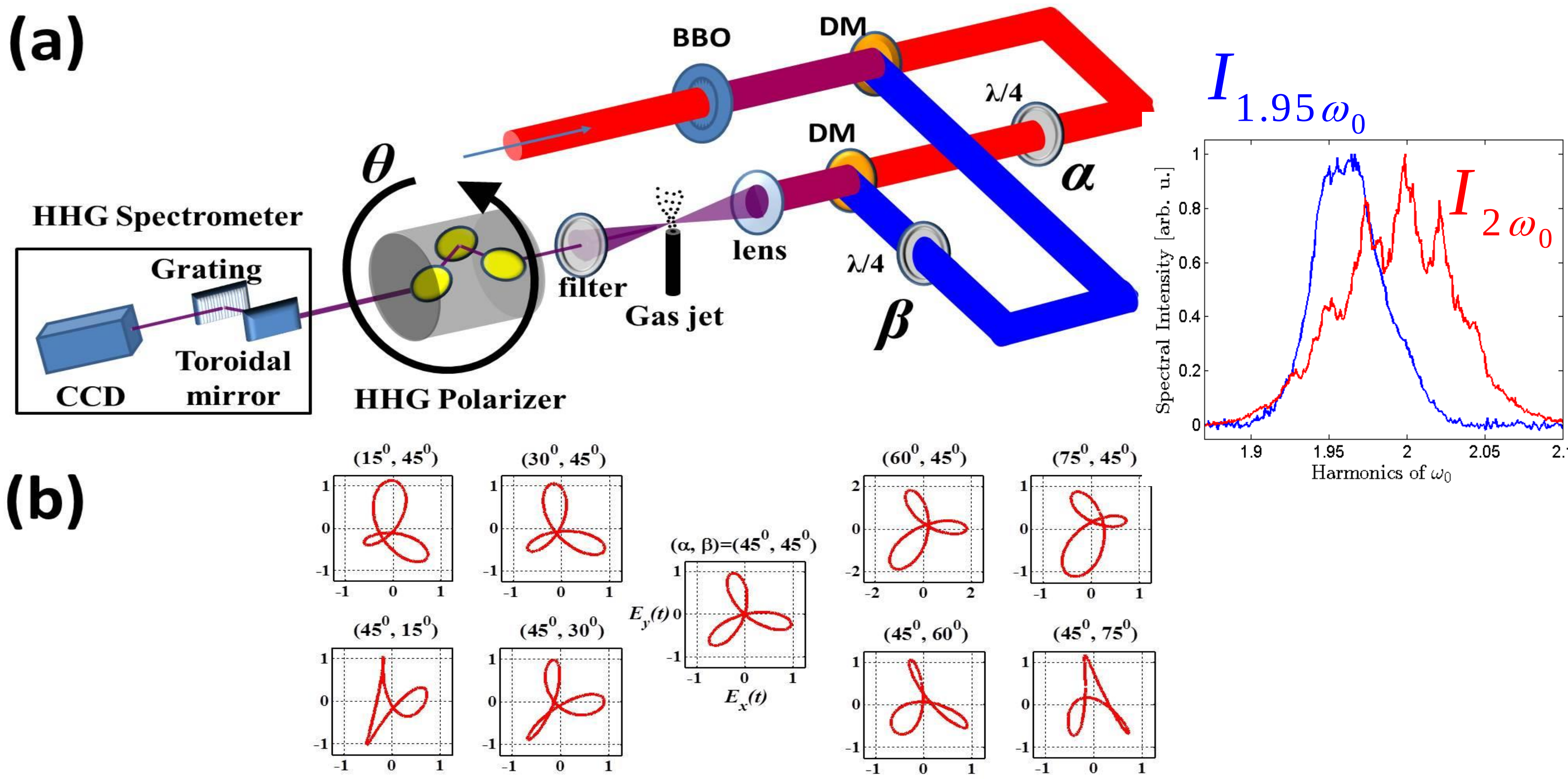
- **Sub-cycle synchronization of 3 recollisions**

$$\begin{cases} a_x(t) = a_1(t) + \cos\left(120^0\right) a_1\left(t - \dfrac{T}{3}\right) + \cos\left(240^0\right) a_1\left(t - \dfrac{2T}{3}\right) \\ a_y(t) = 0 + \sin\left(120^0\right) a_1\left(t - \dfrac{T}{3}\right) + \sin\left(240^0\right) a_1\left(t - \dfrac{2T}{3}\right) \end{cases}$$

$$\Rightarrow \quad \boxed{\varepsilon_{3k\pm1} = 1}$$

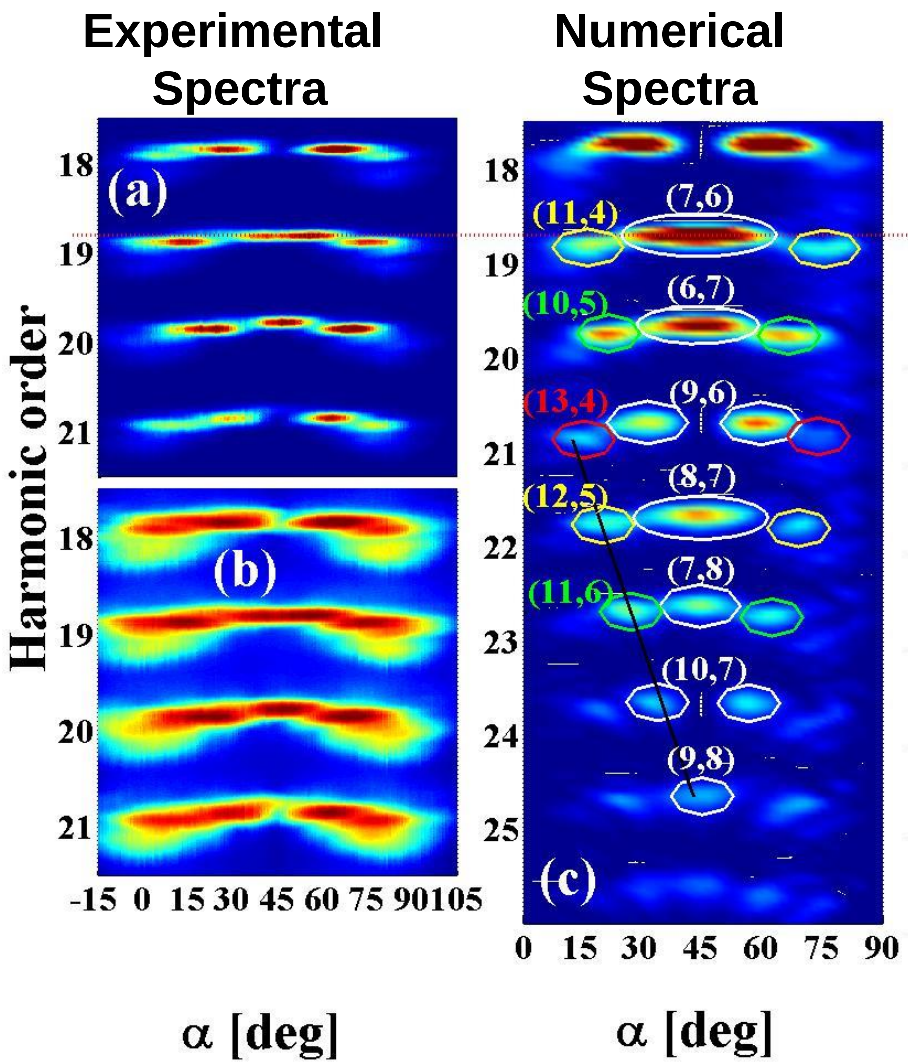


High harmonic generation with counter-rotating elliptically-polarized bi-chromatic fields



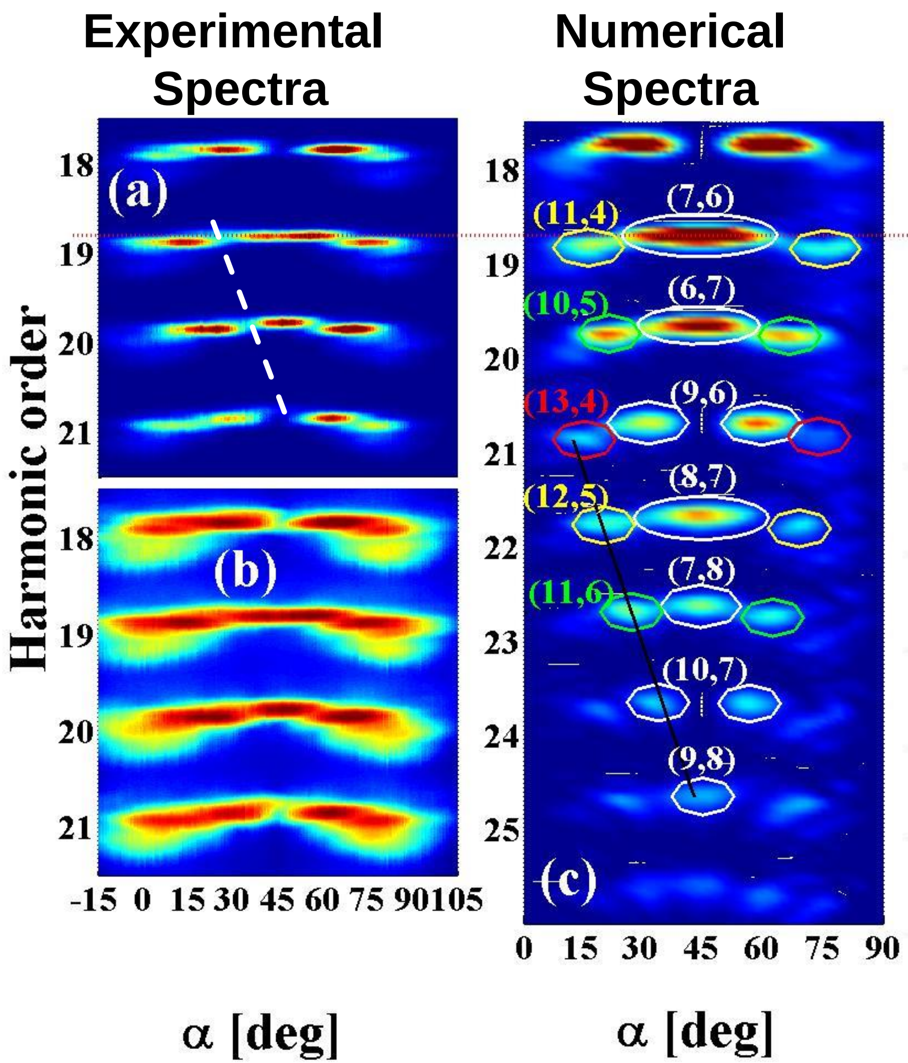
- **Changing α,β .**

Rich spectra – Resolved channels



$$\Omega_{HHG} = n_1 \cdot \omega_1 + n_2 \cdot \omega_2$$

Rich spectra – spin angular momentum



Spin conservation $\left|\sigma\right| \leq 1$

$$\sigma_{HHG} = f(\alpha) n_1 - n_2$$

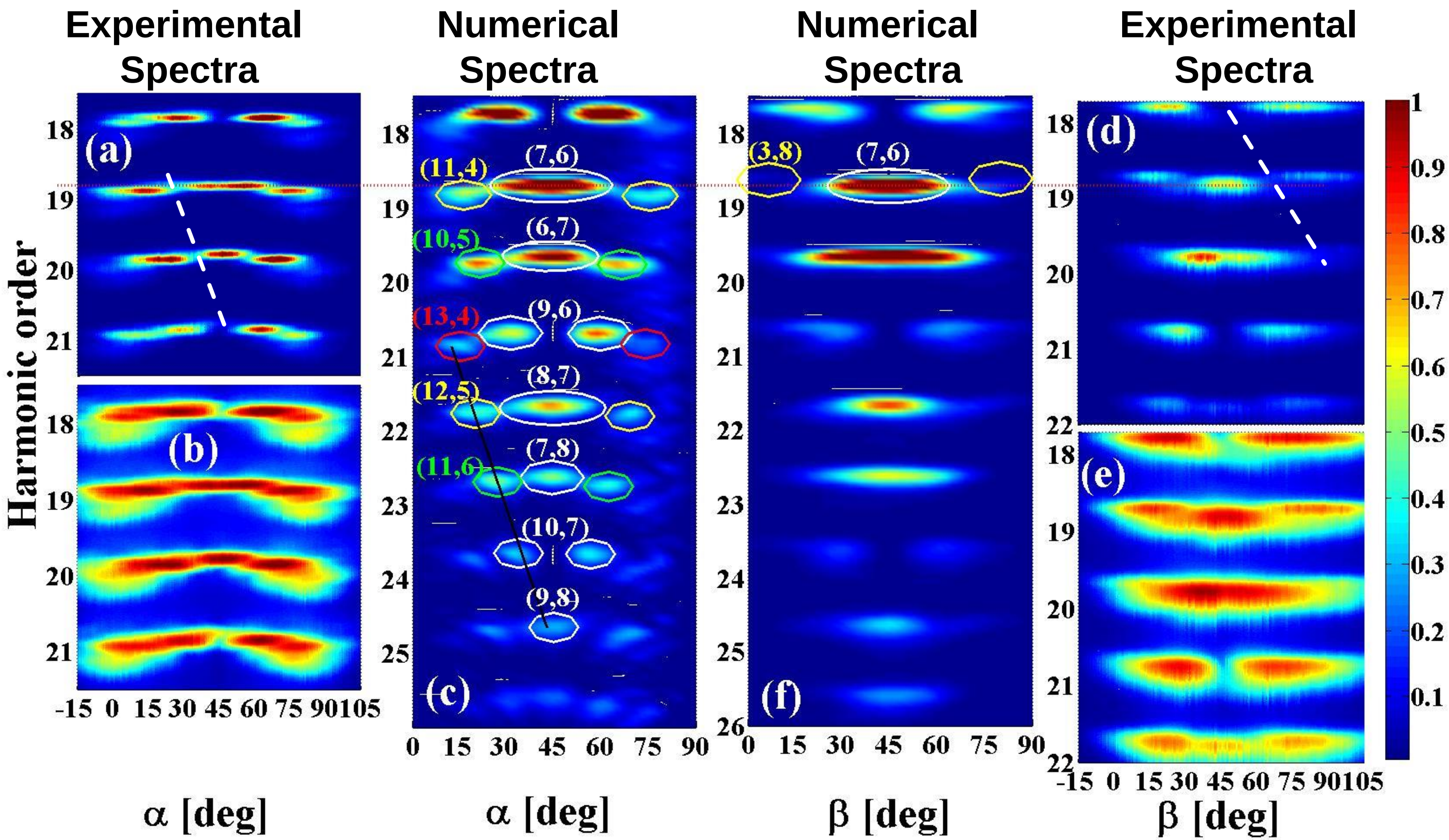
19H: (7 , 6) $f(\alpha) = 1 \rightarrow \sigma_{HHG} = +1$

$f(\alpha) = 5/7 \rightarrow \sigma_{HHG} = -1$

20H: (6,7) $f(\alpha) = 1 \rightarrow \sigma_{HHG} = -1$

$$\Omega_{HHG} = n_1 \cdot \omega_1 + n_2 \cdot \omega_2$$

Rich spectra – spin angular momentum



$$\Omega_{HHG} = n_1 \cdot \omega_1 + n_2 \cdot \omega_2$$

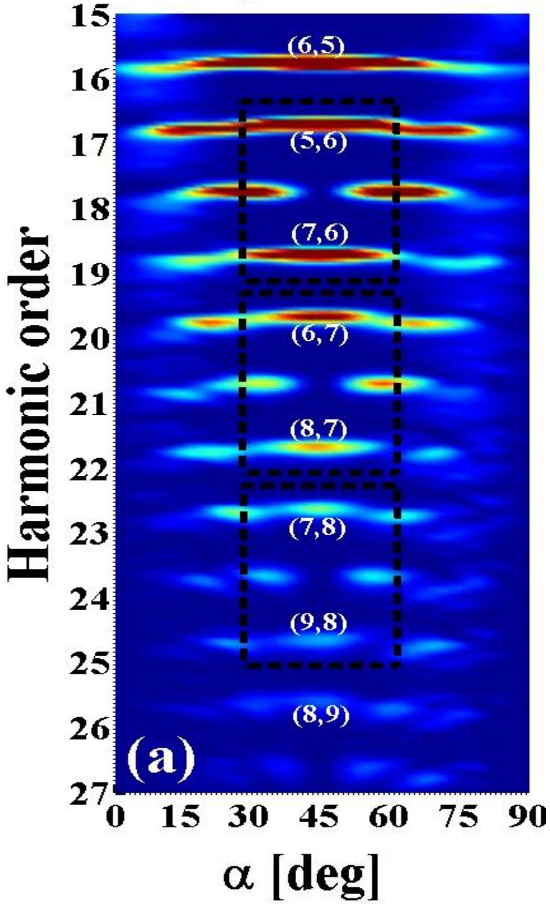
$$|\sigma|, |f(\alpha)|, |g(\beta)| \le 1$$

$$\sigma_{HHG} = f(\alpha) n_1 - n_2$$

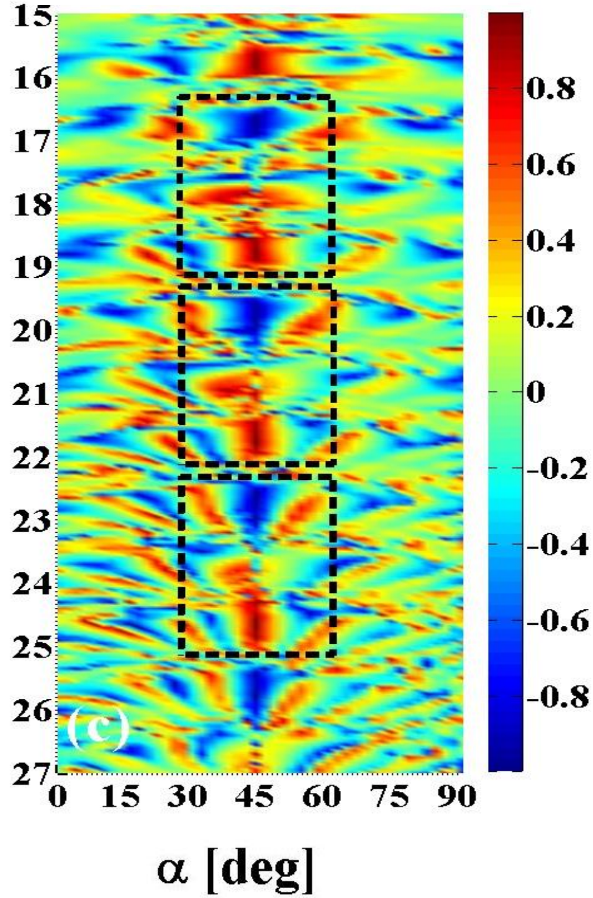
$$\sigma_{HHG} = n_1 - g(\beta) n_2$$

Controlling HHG ellipticity

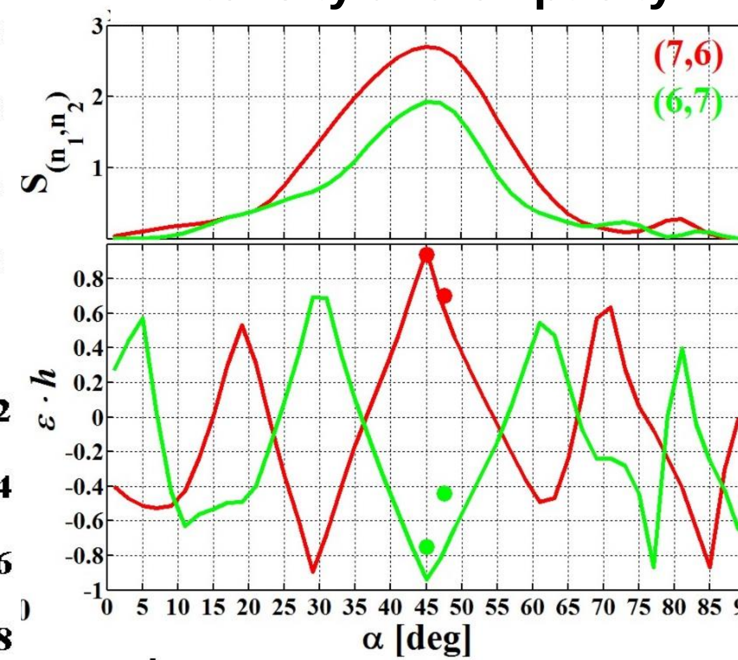
Numerical spectra



Numerical ellipticity * helicity



Experimental intensity and ellipticity



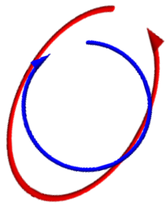
- **Changing α by as little as 8^0 modifies the polarization of H19 from circular (ε=1) to linear (ε=0).**

Discrepancy in conservation of spin angular momentum

- **Energy conservation:** $\Omega_{(n_1,n_2)} = n_1 \cdot \omega + n_2 \cdot 1.95\omega$

$$\Updownarrow$$

- **Spin conservation:** $\sigma_{(n_1,n_2)} = n_1 \cdot \sigma_1 + n_2 \cdot \sigma_2 \;, \; |\sigma| \le 1$

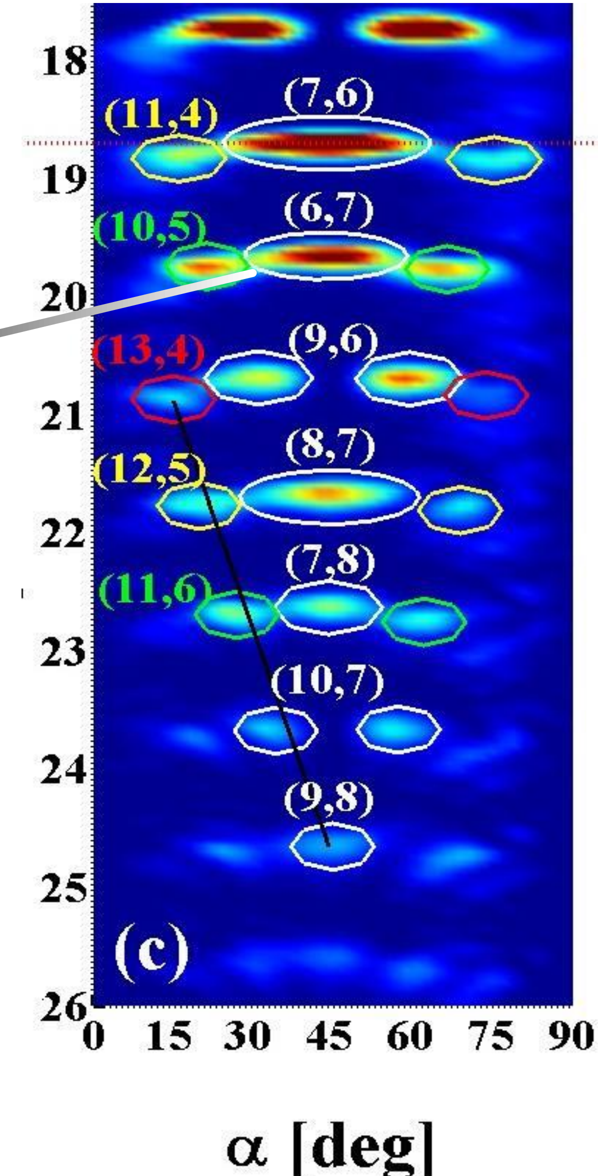


$$\Omega_{H19.65} = 6 \cdot \omega + 7 \cdot 1.95\omega$$

$$\sigma_{H19.65} = 6 \cdot \sigma_1 + 7 \cdot (-1)$$

$$\sigma_1 = \begin{cases} 1 & \alpha = 45^0 \\ 1, \cancel{-1} & \alpha \ne 45^0 \end{cases}$$

$$|\sigma_{H19.65}| \le 1 \quad \Rightarrow \quad \boxed{\sigma_{H19.65} = -1 \quad \forall\, \alpha}$$



- **H19.65 should remain circularly-polarized regardless of the value of α.**
- **Experiment and numerics indicate the opposite!**

Additional (radiation or electronic) quanta

Possible solutions:

- **Conservation law hold true for harmonic pairs:**

$$\Omega_{(n_1,n_2)} + \Omega_{(n_2,n_1)} = (n_1 + n_2)\cdot\omega + (n_1 + n_2)\cdot 1.95\omega$$

$$\sigma_{(n_1,n_2)} + \sigma_{(n_2,n_1)} = (n_1 + n_2)\cdot\sigma_1 + (n_1 + n_2)\cdot\sigma_2$$

→ **quantum optics**

- **Medium transfers angular momentum**
  - **Strong-field → anisotropic media**

  → **Process is not parametric**

  → **HHG spectroscopy of circulating current @ attosecond resolution**

Summary

- **Full control over polarization of HHG**

  - by attosecond & angle control over the 2D recollissions

- **Role of spin angular momentum in extreme NLO**

- **Resolve (n_1,n_2) channels using single-atom physics**

- **Conservation of spin angular momentum:**

- Qualitative agreement with experimental & numerical results

- Quantitative disagreement with experimental & numerical results

→ missing quanta. Radiation or electronic?

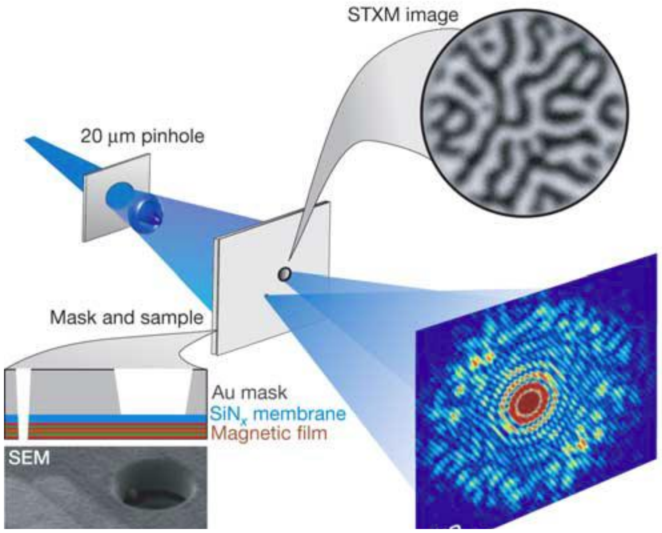
Next…

- High spatiotemporal imaging of magnetic domains.

- Molecules

- Attosecond pulses with circular and elliptic polarization

- Phase Matching



Eisebitt, S., *et al.*, Nature **432**, 885 (2004)
(by synchrotron)