

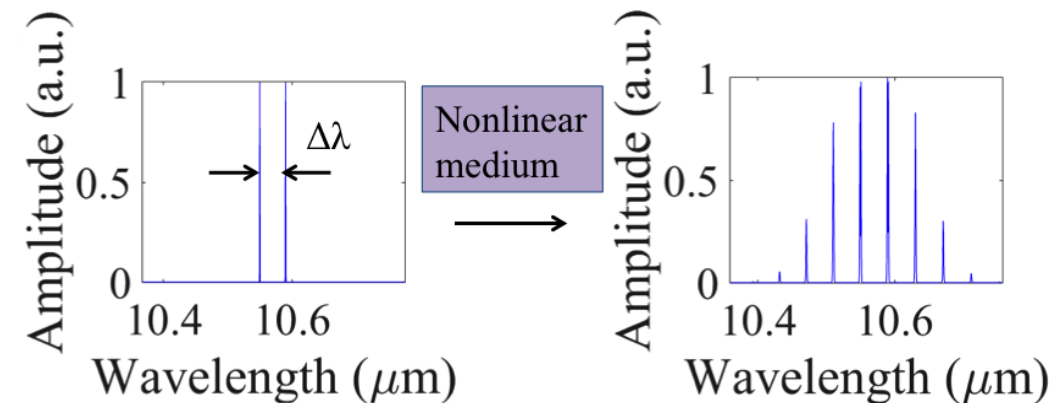


Science and
Technology
Facilities Council

Laser harmonic generation in plasma: the new beat wave

Raoul Trines, Holger Schmitz, Bob Bingham
Central Laser Facility
Rutherford Appleton Laboratory

Martin King, Paul McKenna
Dept. Physics, U. Strathclyde, Glasgow



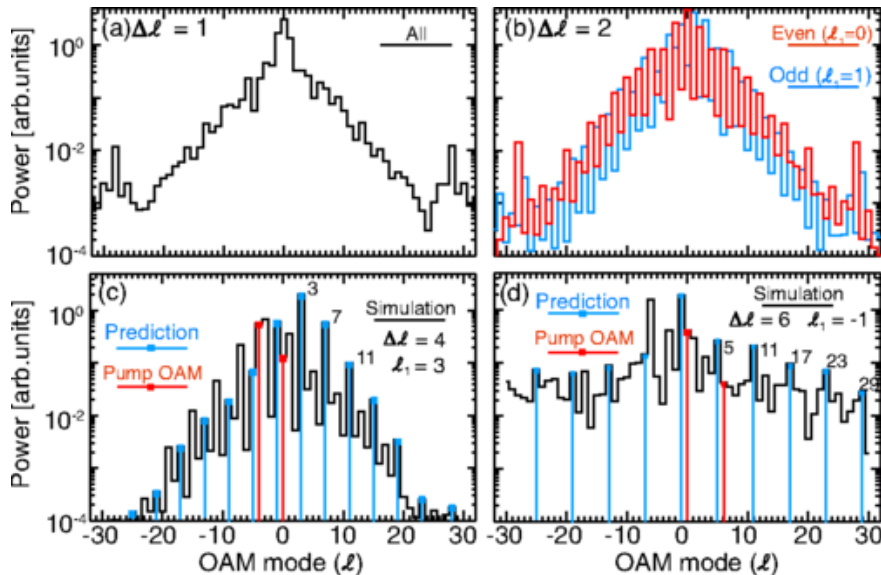
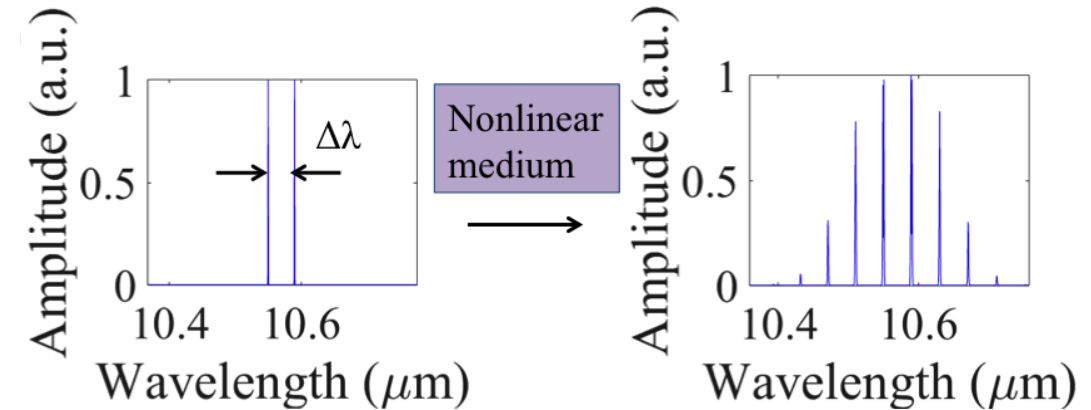
Summary

- Describe high harmonic generation as a beat wave process with beat wave spectra
- Novel analysis using CP waves, signed frequencies and $\Psi = E_x + i^*E_y$
- Use CP laser beams and structured targets to generate spectra “to order”
- Generate bespoke frequency combs by taking a 1-D subset from a 2-D spectrum
- Describe the HHG spectrum via symmetries of the beam-target configuration
- Relate to spectral theorems by Noether, Laue, Mathieu, Floquet and Bloch

Beat wave to drive plasma waves

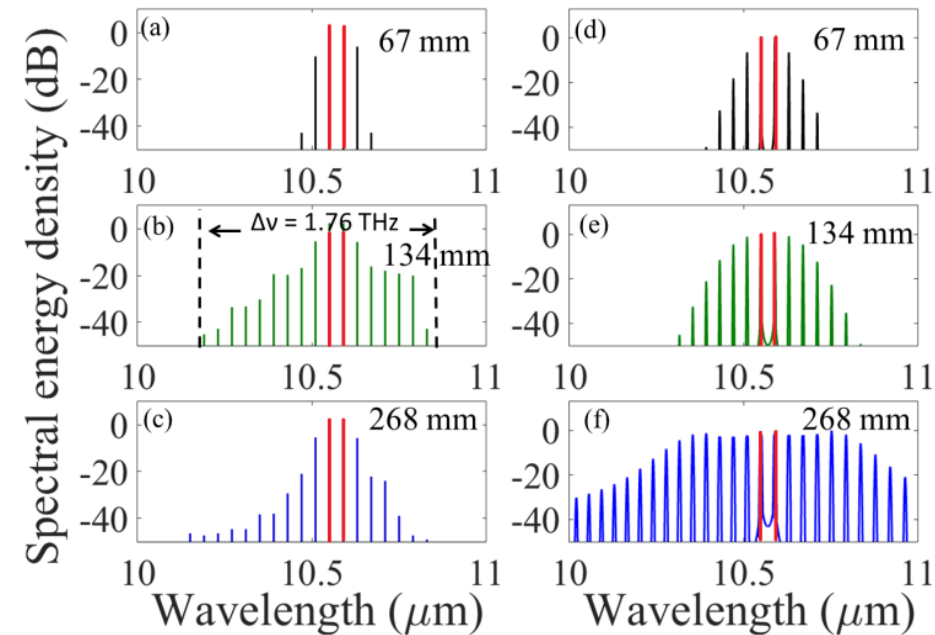
- Start with two spectral lines with separation $\Delta\lambda$ or $\Delta\omega$
- Let those “beat” in a (resonant) nonlinear medium
- Perturbation in medium with frequency $\Delta\omega$: plasma wave to accelerate electrons
- Spectrum shows “side bands”: extra lines also separated by $\Delta\omega$
- Similar: **OAM side bands** in Raman amplification

• All harmonic spectra as beat waves !?



J. J. Pigeon, S. Ya. Tochitsky, and C. Joshi, Optics Letters 40, Issue 24, 5730 (2015)

J. Vieira, R. Trines et al., Phys. Rev. Lett. 117, 265001 (2016).

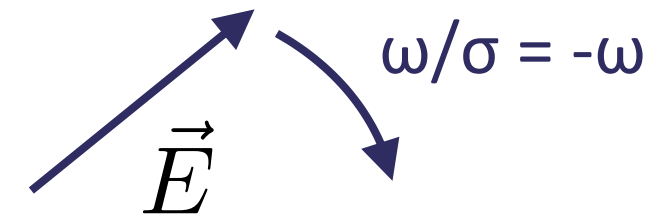
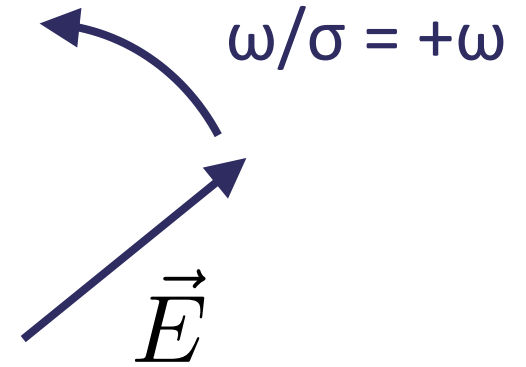


From beat wave to HHG: conventions used here

Need to use special conventions to describe a harmonic spectrum as a jumped-up beat wave

- Write all laser fields as **superpositions of CP laser modes** (yes, the opposite of what everyone else does!)
- Spin $\sigma = \pm 1$, frequency $\omega > 0$ always
- Treat LCP and RCP modes as **different harmonics** with different signs of ω/σ rather than two polarisations of the same harmonic

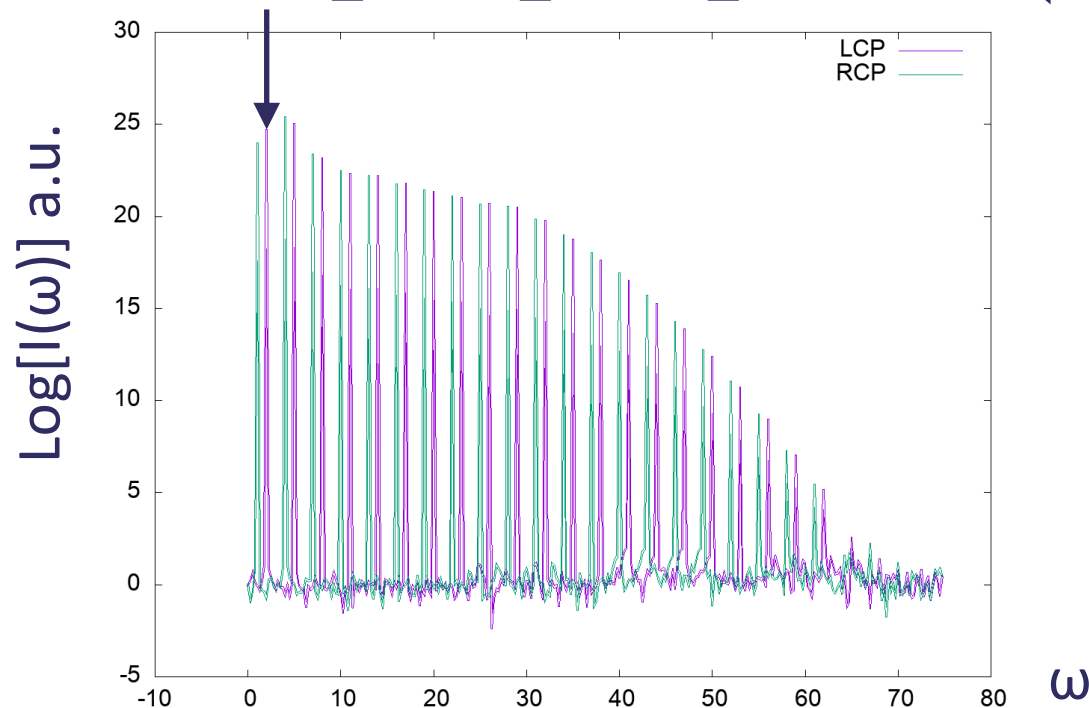
These conventions will make it much easier to calculate and visualise the harmonic progression (as a beat wave)



Why ω/σ ? Example: PRA 98, 011401 (2018)

- Pump: ω with RCP, 2ω with LCP, $\Delta\omega=1$, $\Delta(\omega/\sigma)=3$
- ω -spectrum stutters, some frequencies "suppressed"
- ω/σ -spectrum is regular, looks like a **beat-wave** spectrum

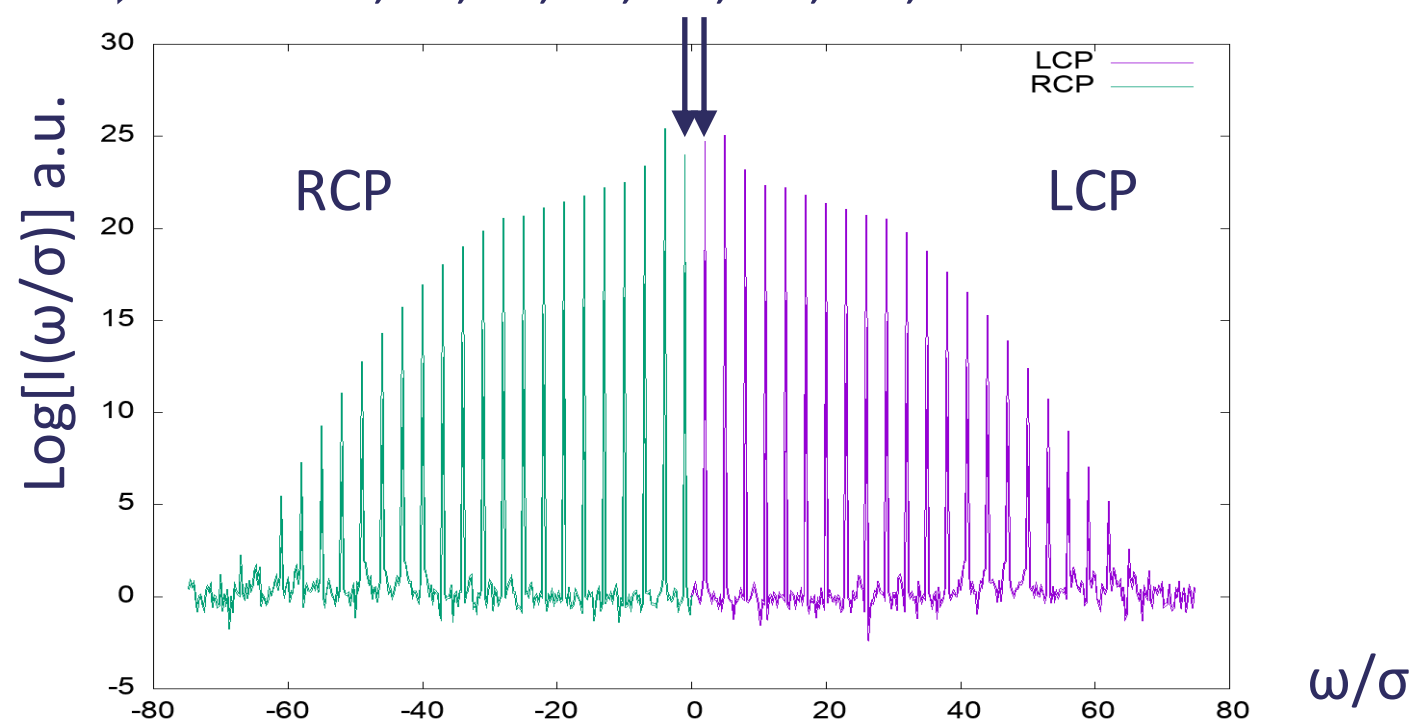
ω -spectrum from paper
1, 2, _, 4, 5, _, 7, 8, _, ...



Same data!



ω/σ -spectrum by me
..., -8, -5, -2, +1, +4, +7, ...



Two variables: frequency ω , OAM level ℓ

- Two pump modes with frequencies ω and 2ω , OAM levels +1 and +1, and L- and R-circular polarization
- $\Delta\omega=1$, $\Delta\ell=0$, does not match step sizes in graph
- Need two curves to describe L- and R-polarized harmonics separately
- Relationship between pump modes at (1,1) and (2,1) and harmonic progression not obvious

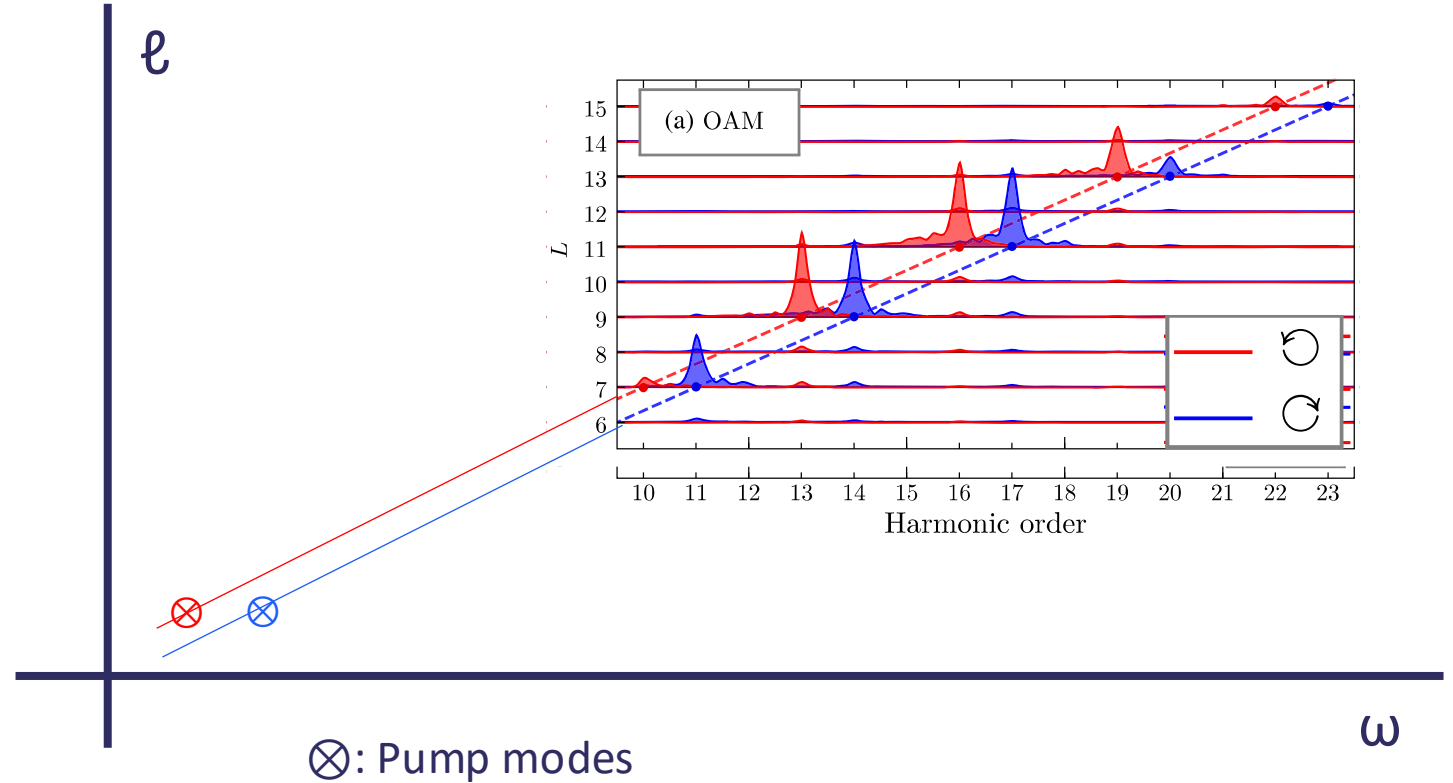


Image from PRL **122**, 203201 (2019).

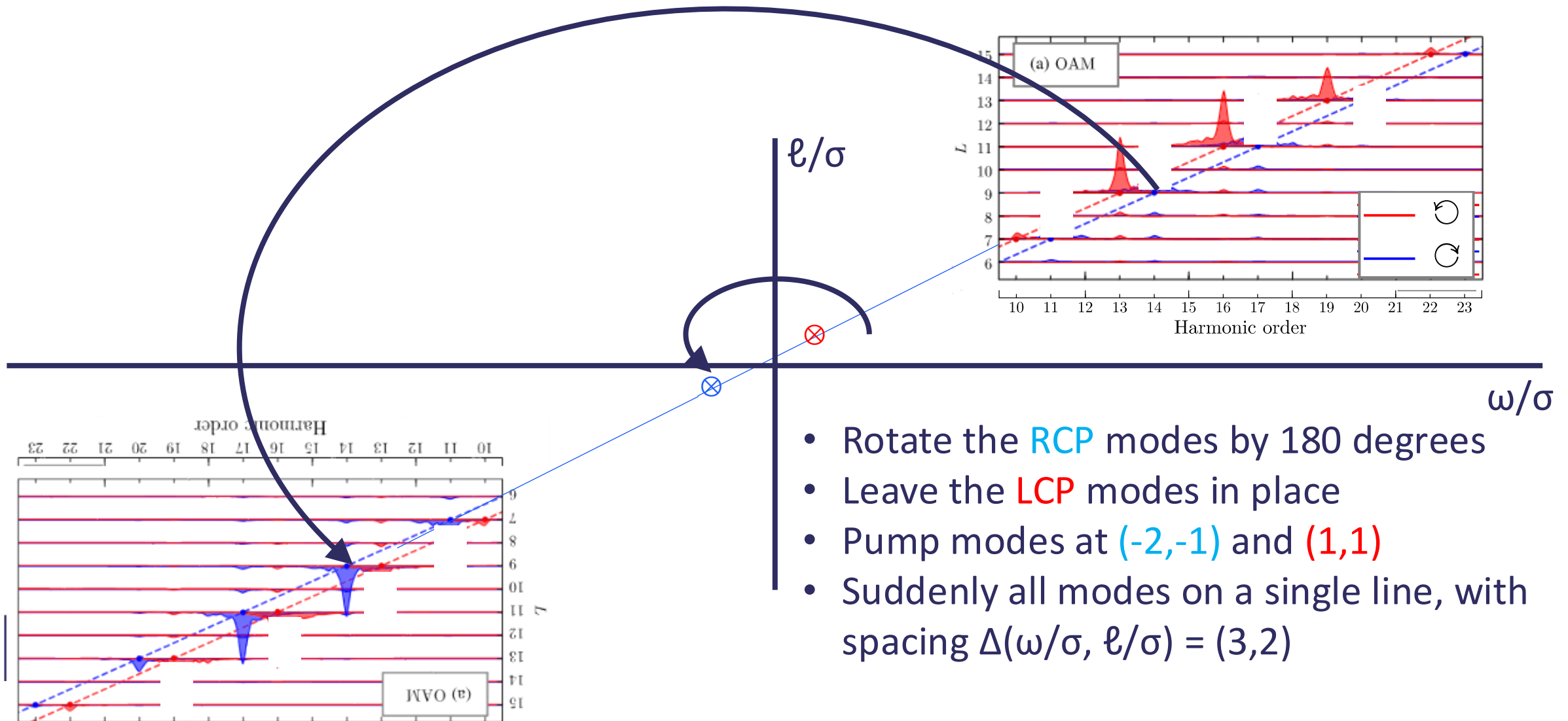
E. Pisanty et al., Phys. Rev. Lett. **122**, 203201 (2019).

K. Dorney et al., N. Photon **13**, 123 (2019).

B. Minneker et al., Phys. Rev. A **104**, 053116 (2021).

D. Xie et al., Front. Phys. **10**, 962956 (2022).

Now use ω/σ and ℓ/σ instead of ω and ℓ

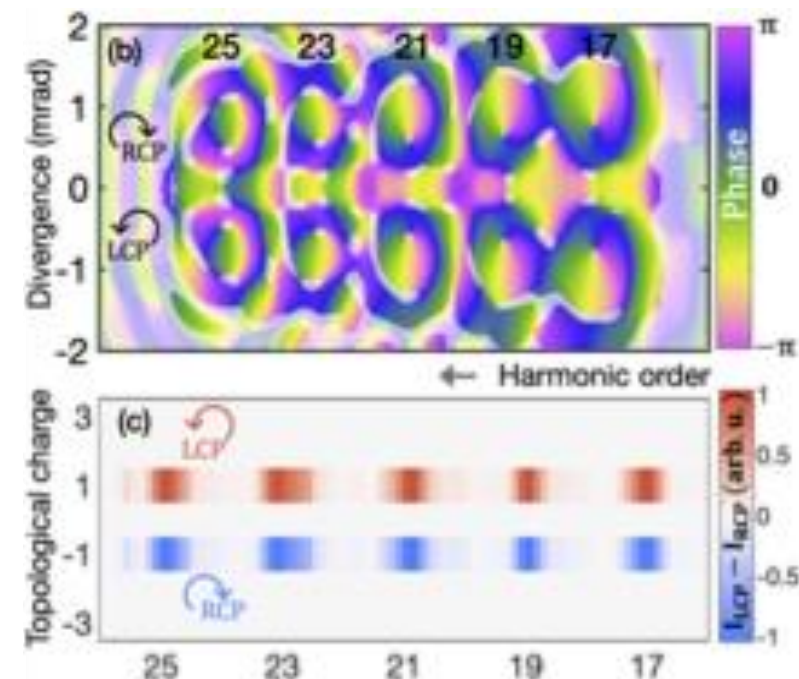
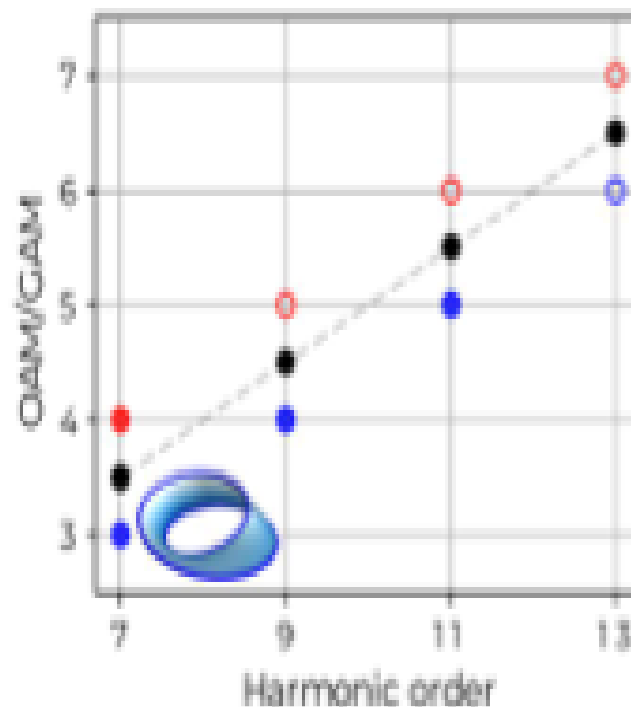
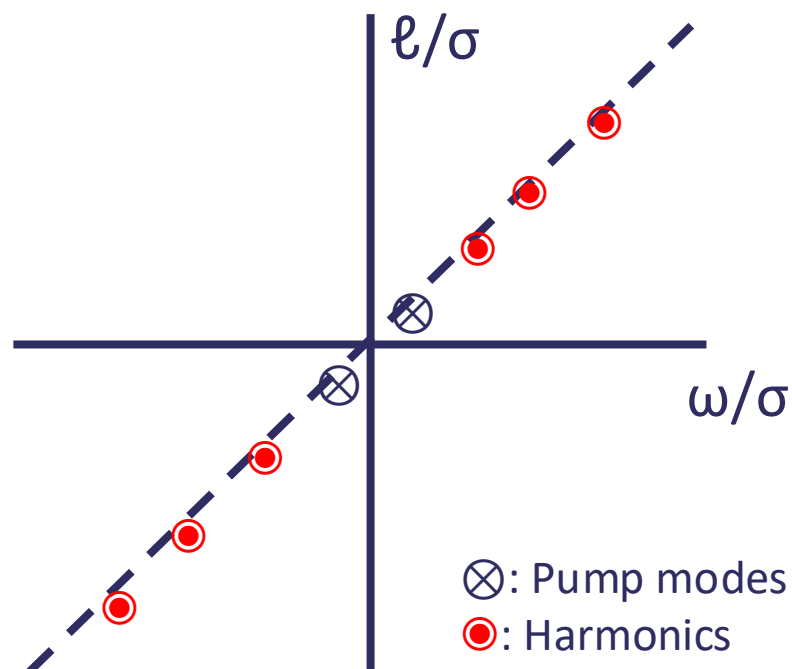


HG with funky laser beams

LP beam with OAM, $\ell=1$, $\ell_q = q*1$
Zhang *et al.*, PRL **114**, 173901 (2015).

Möbius beam, Luttmann *et al.*,
Sci. Adv. **9**, eadf3486 (2023).

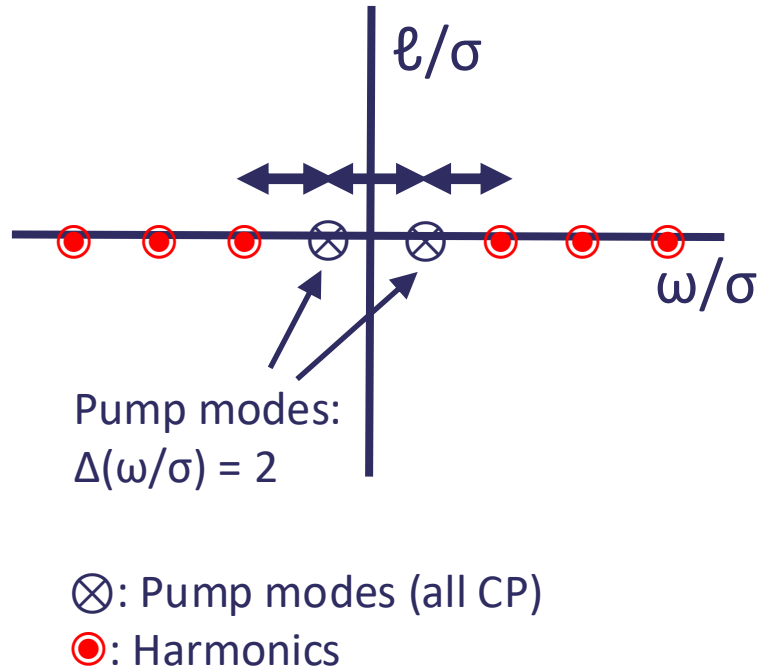
Beam with radial polarisation,
A. De Las Heras *et al.*, Optica
11, 1085 (2024).



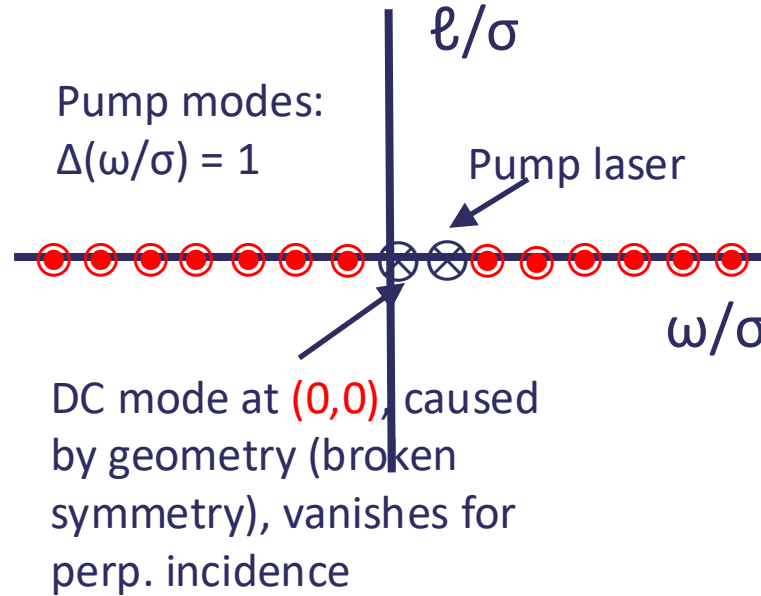
Rotate half the points, then everything
on the same line with equal spacing!

HG in $(\omega/\sigma, \ell/\sigma)$ plots: like a beat wave

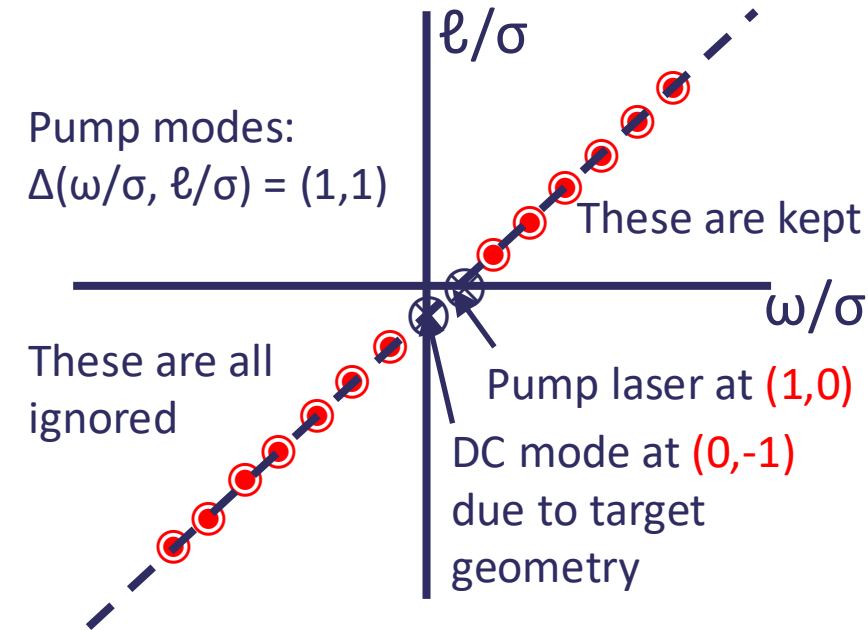
LP, perp. incidence, pump modes at $\omega/\sigma = \pm 1$, only odd harmonics (frequency or OAM)



CP, oblique incidence, all harmonics thanks to "DC mode" at $\omega/\sigma = 0$

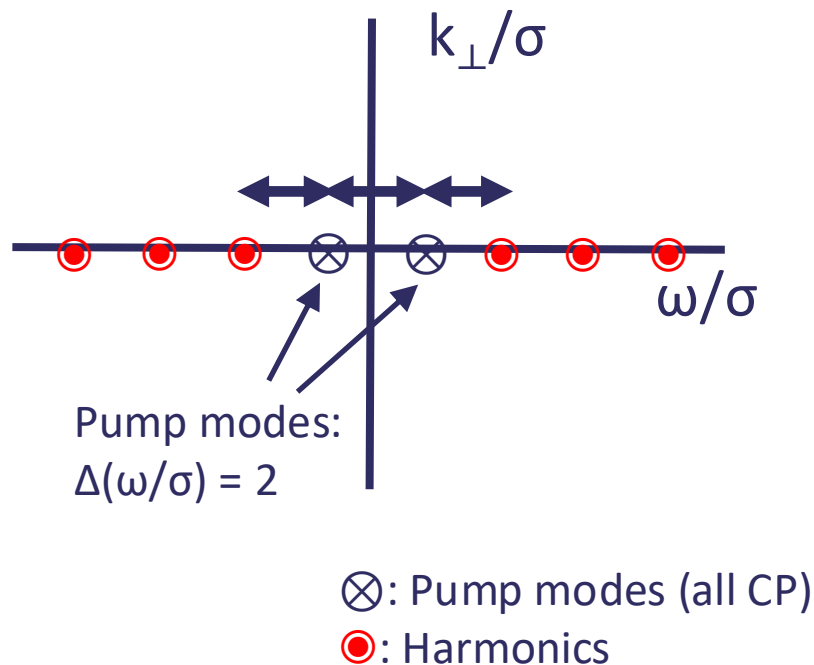


CP without OAM, tight focus onto circular dent in target "Negative" harmonics always ignored



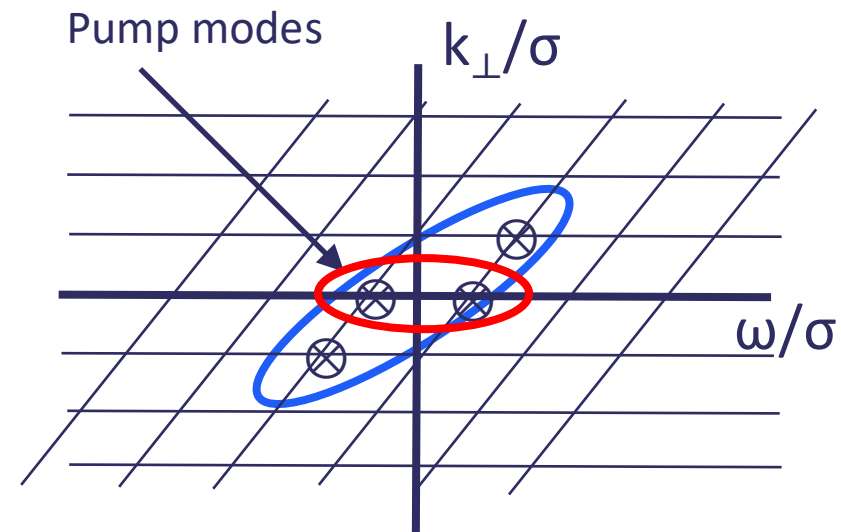
Harmonic progressions: 1-D vs 2-D

One LP pulse, 1-D progression,
pump modes at $\omega/\sigma = \pm 1$, only
odd harmonics



Two LP pulses at ω and 2ω

- Four modes, 2-D grid
- Many values of $k_{\perp}$ per frequency ω
- Spectrum is inversion-symmetric, so all modes recombine to LP harmonics



Shape your target: Lichters' model revisited

- **Lichters model**: intense laser pulse hits flat solid target. **Oscillating** target surface drives **harmonics** in the reflected light
- Lichters' model: "oblique incidence" → vertical incidence on **tilted target**
- Describe target surface with "height function" $h(x,y)$
- Lichters' equation revisited ($\Psi = E_x + i^*E_y$):

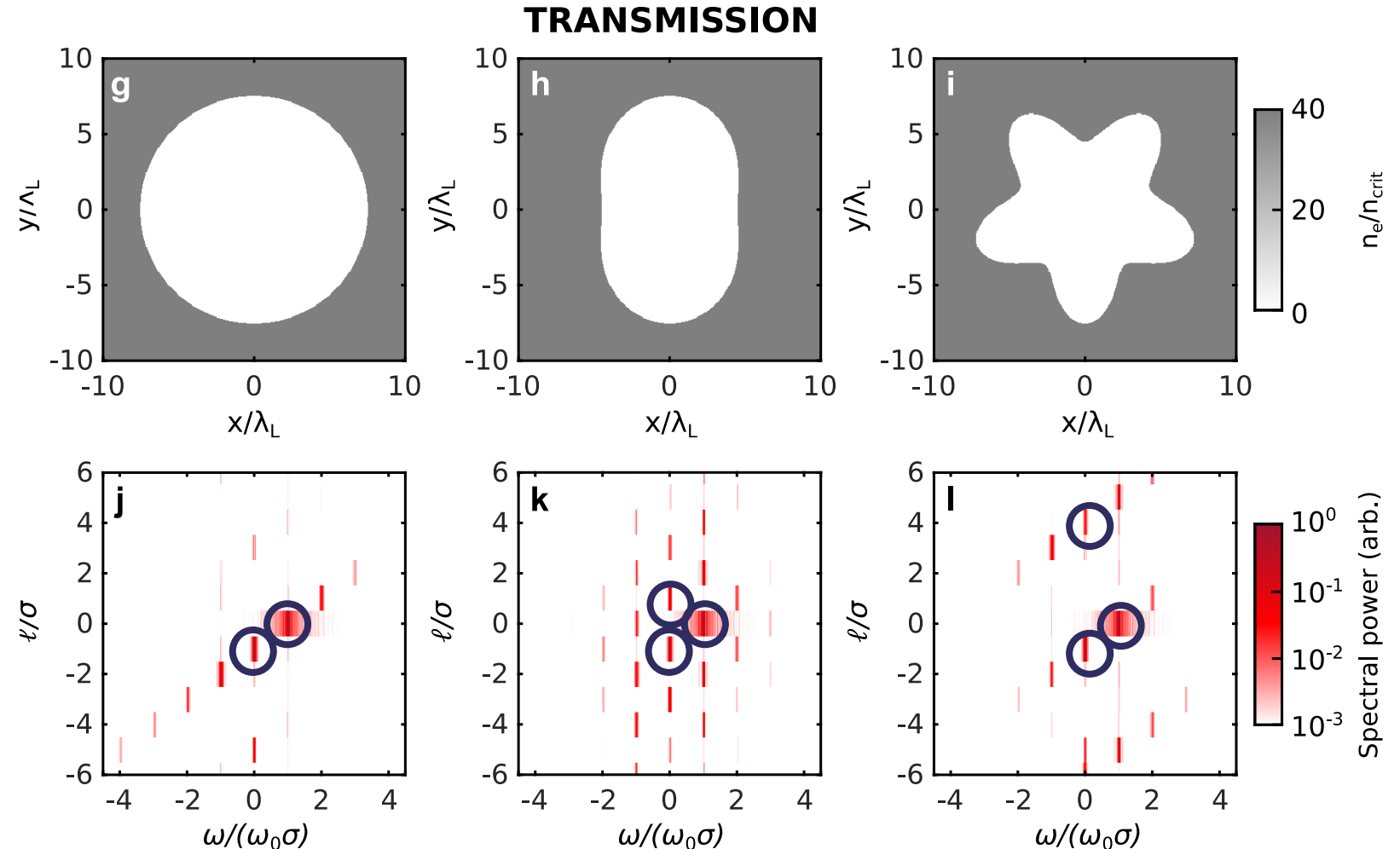
$$(\partial_t^2 - c^2 \nabla^2) \Psi = -\omega_p^2 (1 + \Psi^* \Psi + \Psi \cdot \nabla h) \Psi$$

- Ψ **beats** with ∇h , so ∇h provides the "DC mode" with zero ω but **nonzero ℓ/σ**
- **Shape your target** to determine **ℓ/σ** of the "DC mode"
 - Tilted flat target (Lichters et al.): $\nabla h = \tan(\alpha) e_x$, **$\ell/\sigma = 0$**
 - Circular dent or aperture (Wang et al., Li et al., Yi): $\nabla h \propto e_r$, **$\ell/\sigma = -1$**
 - General "q-plate" (our work): $h = r^{1-q} \cos[(1-q)\varphi]$, **$\ell/\sigma = -q$**

Simulation results: aperture targets (**q=1, 2, 5**)

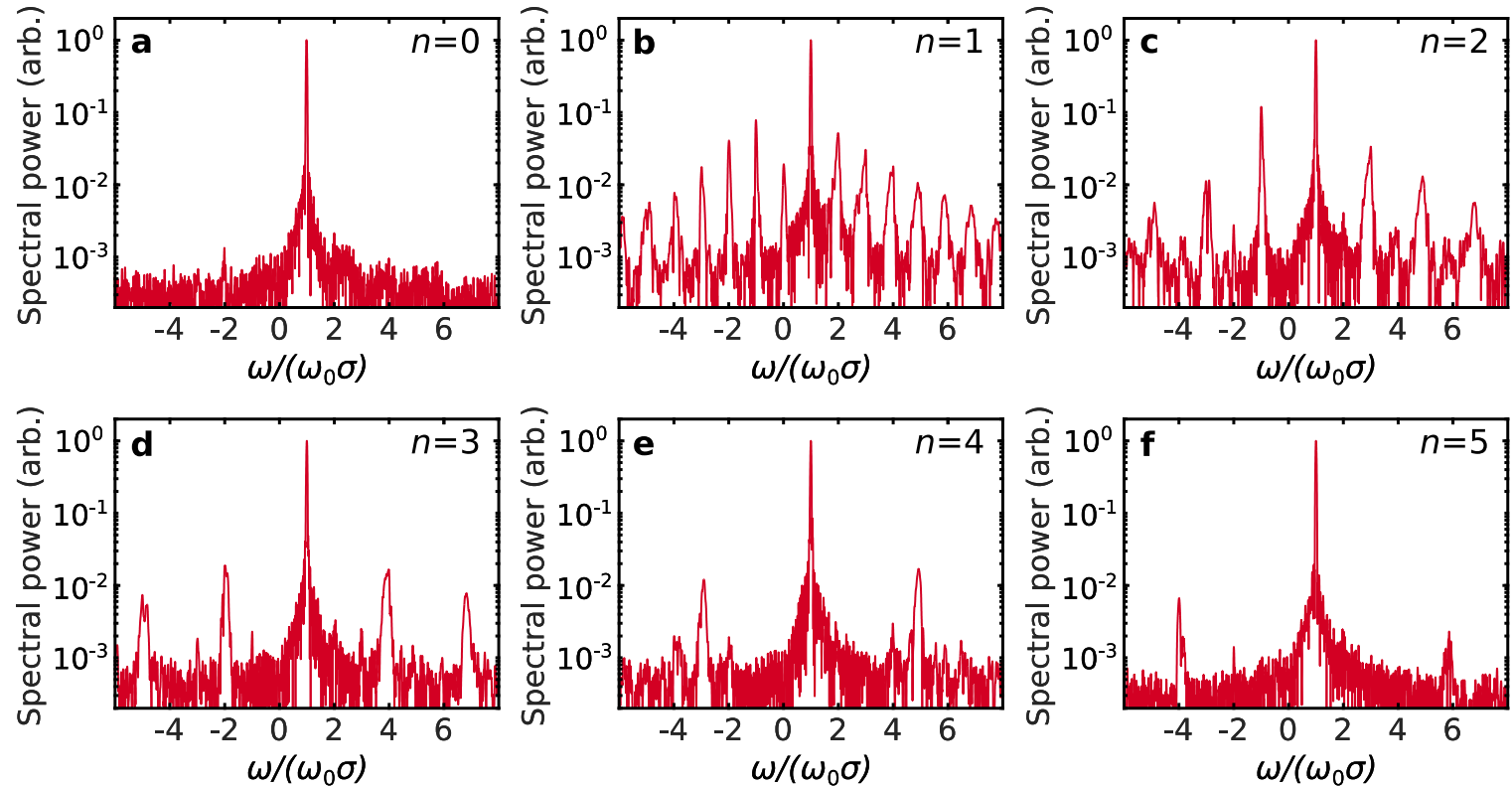
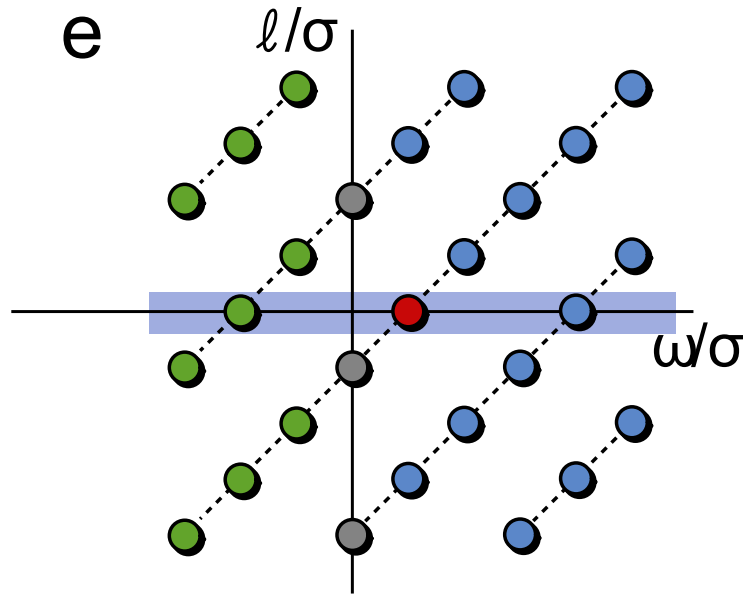
Simulations and data processing: H. Schmitz, M. King

- Calculate spectrum of $\Psi(\varphi, t) = E_x + i^*E_y$
- Pump laser mode and target modes **ringed**
- Circular target provides one mode, **1-D grid**
- Other targets provide two modes, **2-D grid**
- Result: always the regular grid **generated by the ringed modes**



Frequency comb: 1-D subset from 2-D spectrum

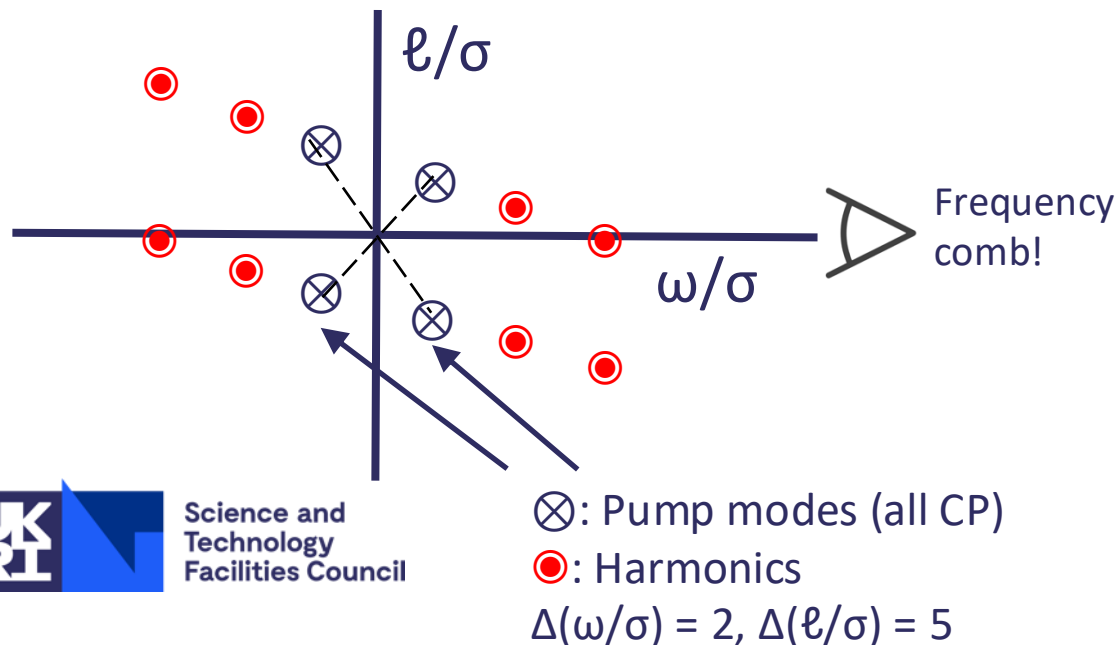
- From earlier 2-D spectrum: select on-axis light to select modes with $\ell/\sigma=0$
- Only selected frequencies have an $\ell/\sigma=0$ mode: frequency comb!
- Shown are combs for circular aperture and apertures with 1-5 fold symmetry
- All 2-D harmonic progressions have the potential of a frequency comb



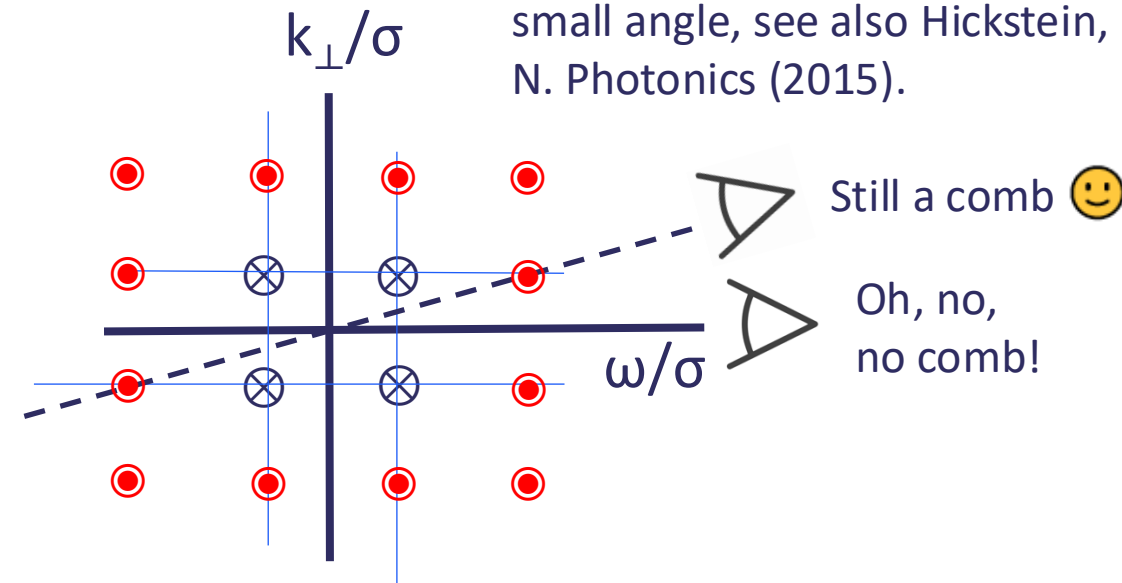
2-D progression: symmetric frequency comb

- “Necklace beam”, Rego et al., Sci. Adv. (2022): two LP beams with “nearly opposite” OAM, progression in (ω, ℓ) space, **symmetric** comb on $\omega=0$ axis
- Hickstein, N. Photonics (2015): two LP beams at small angle, progression in $(\omega, k_{\perp})$ space, but **no mention of combs**, and no harmonics **on-axis**
- Our idea: start from earlier work, but look **off-axis** for comb in $(\omega, k_{\perp})$ space

L. Rego et al., Sci. Adv. **8**, eabj7380 (2022).



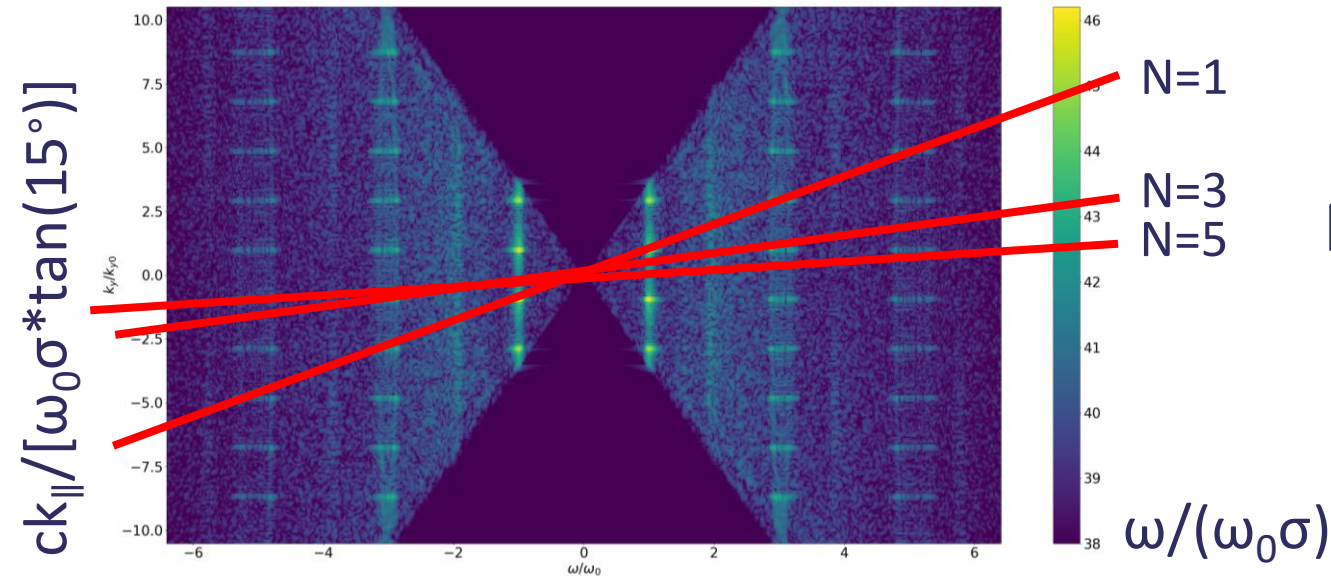
Two LP beams, separated by a small angle, see also Hickstein, N. Photonics (2015).



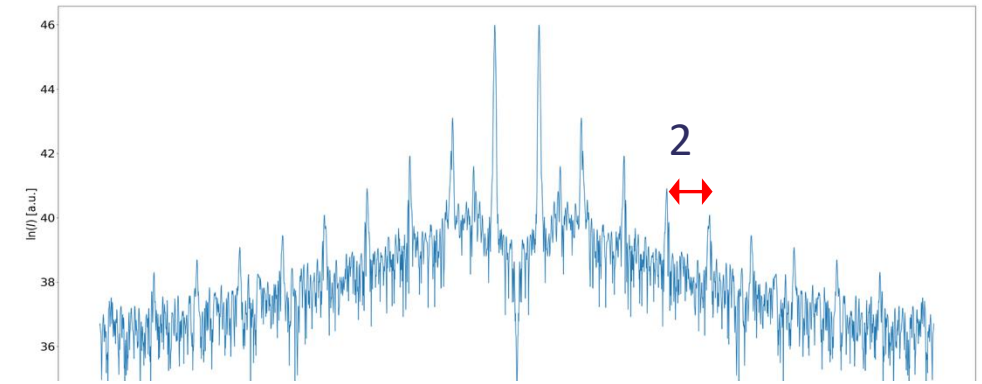
Hickstein: **angles** for given **frequency**
Here: **frequencies** for given **angle**

Simulation results: super-Gaussian pulses

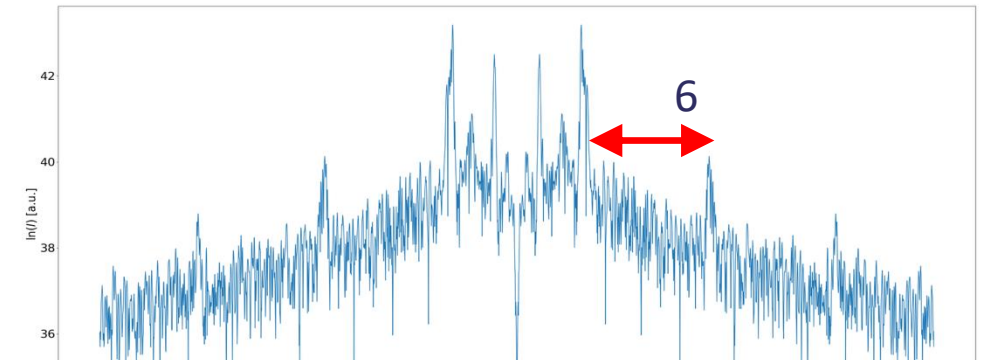
- Spectral cuts: $ck_{\perp}/[\omega \sigma \tan(15^\circ)] = 1, 1/3, 1/5$
- Peaks at $(2n+1)*1, (2n+1)*3, (2n+1)*5$,
from the same simulation!
- A range of combs can be selected, depending on your viewing angle
- Simple, versatile, easily extended



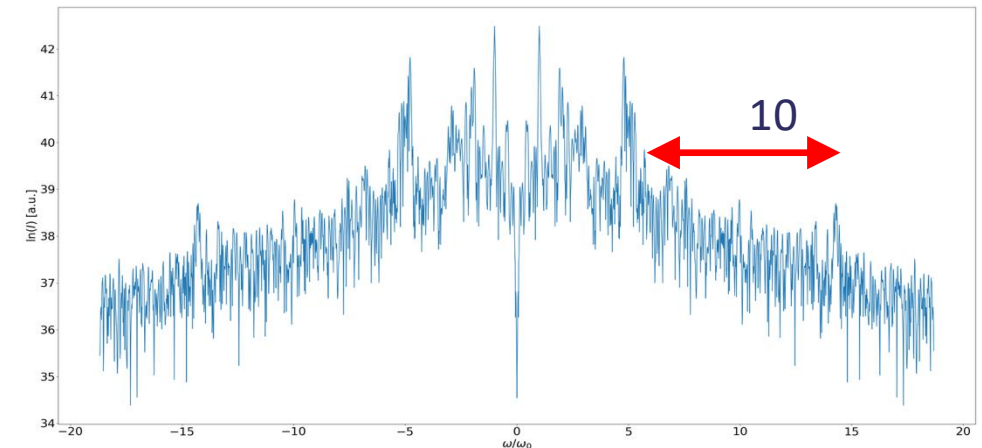
N=1



N=3

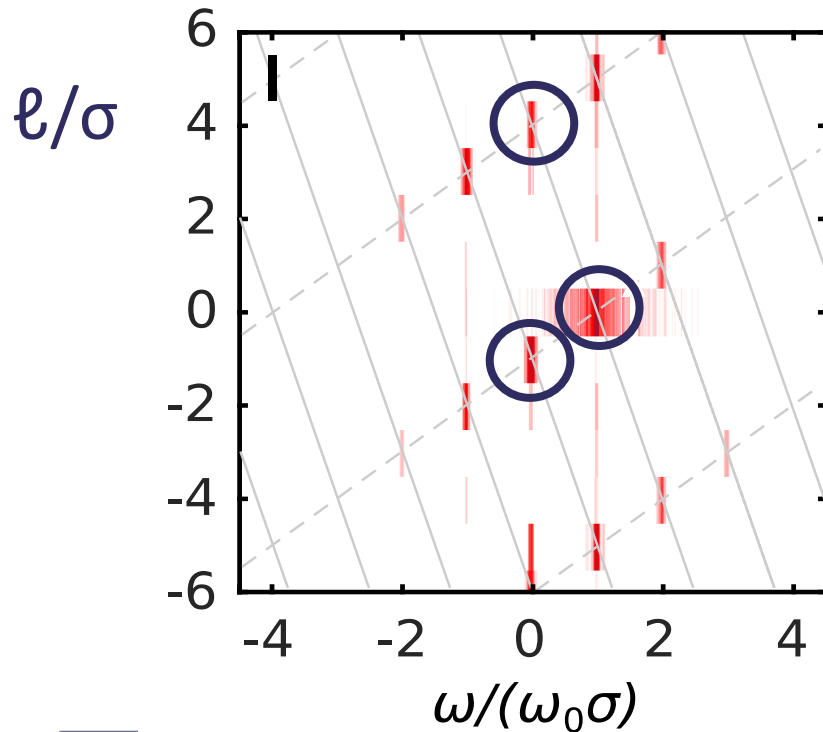


N=5

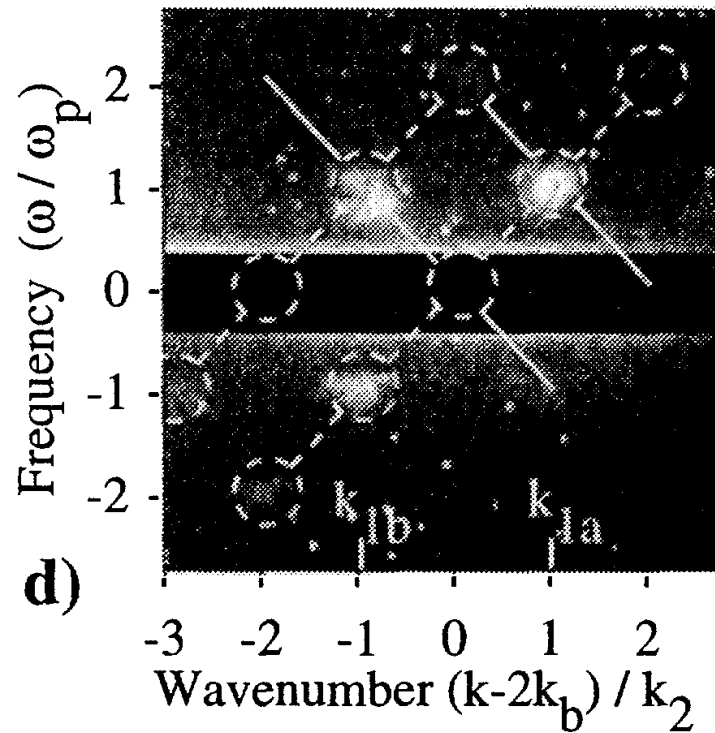


HHG vs beat wave vs crystal scattering

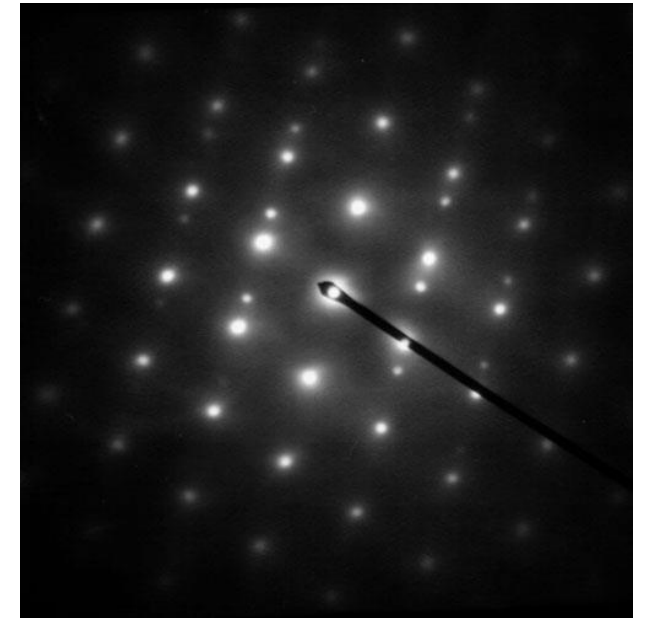
Our work on HHG:
NComms **15**, 6878 (2024)



Old beat wave result:
PRL **74**, 2236 (1995)



Electron scattering off a crystal (Wikipedia)

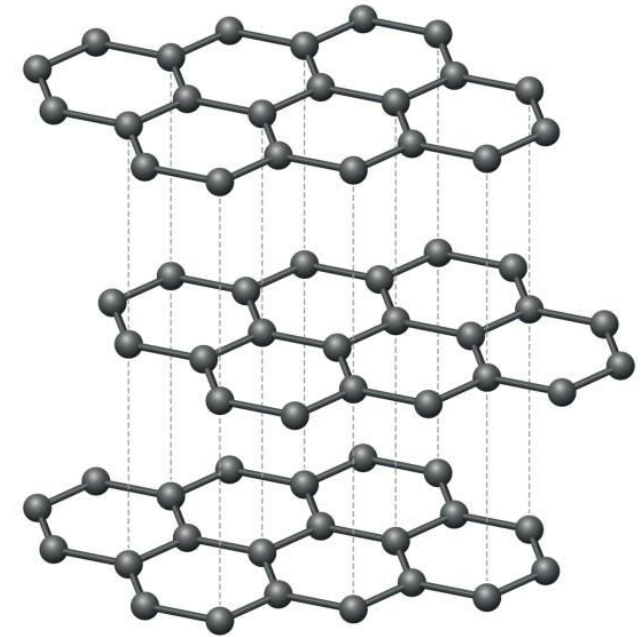


Similar spectra, similar underlying theory!?

Symmetries?

Symmetry: an isometric coordinate transformation (translation, rotation, mirror, inversion) that **leaves a system invariant** and **maps solutions onto solutions**

- Much used in **crystal theory**, to map a crystal onto itself
- Their symmetries **dictate** the wave vector spectrum (**Laue** equations)
- Much less used in the theory of HHG, but **equally relevant**
- Pump laser **satisfies certain symmetries** $\leftrightarrow$ symmetries imposed onto nonlinear term $A^2A \leftrightarrow$ nonlinear term drives harmonics, imposes **the same symmetries** on them



Symmetry theory for HHG: keep A^2 invariant

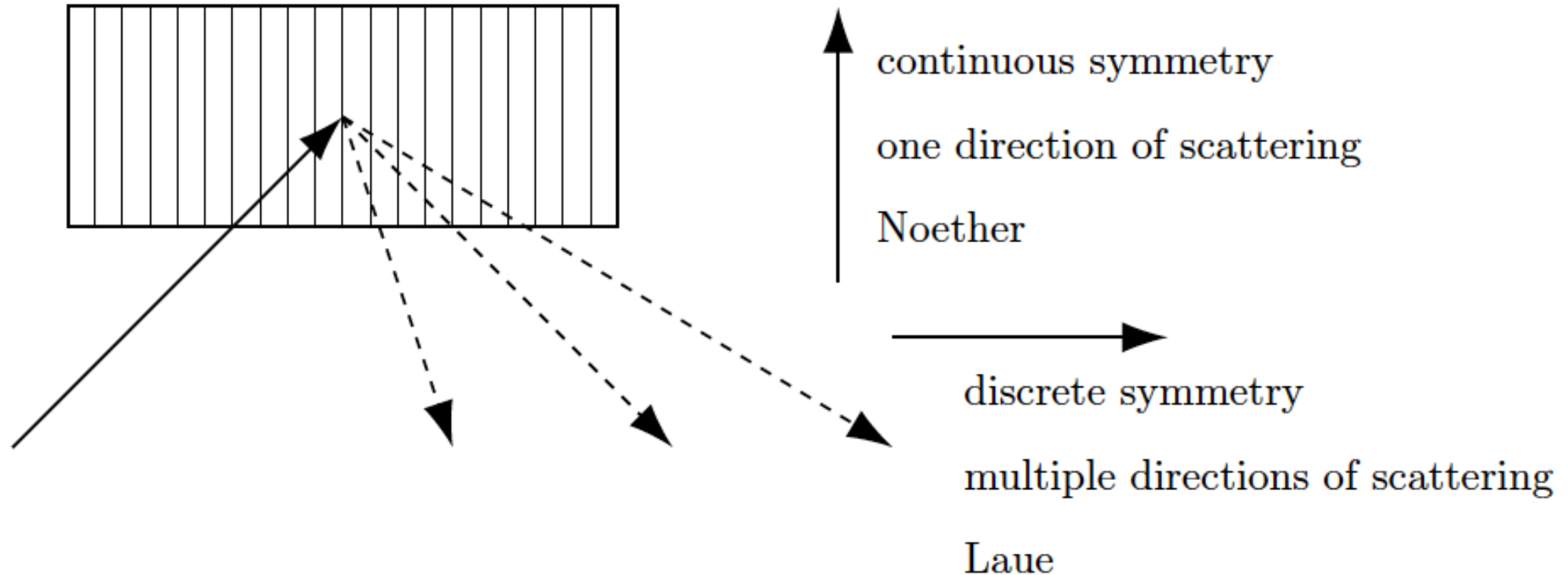
- Marry the **beat-wave** approach to **symmetry theory** and **spectral theorems**
- **Yes**, also important for plasma physics (S. Li et al., NJP **22**, 013054, 2020)
- Describe the HHG spectrum via **symmetries** of the beam-target configuration
- Relate to **spectral theorems** by Mathieu, Noether, Laue, Floquet and Bloch

Symmetries in our HHG model

- Define Hamiltonian $H = \partial_t^2 - c^2 \nabla^2 + f(|\Psi|^2)$, equation $H\Psi = 0$
- Write $\Psi = \Psi_0 + d\Psi$, $\Psi_0 = \Psi_A + \Psi_B + \dots$ (all fundamental modes)
- **Fundamental mode**: any Ψ that leaves **H invariant**, e.g. a CP mode that leaves A^2 (or E^2) constant: “does not beat”; for later; Ψ is an **eigenfunction for coordinate translations**
- Evolution of $d\Psi$: $(\partial_t^2 - c^2 \nabla^2)d\Psi + (\Psi_A \Psi_B^* + \Psi_A \Psi_C^* + \dots)\Psi_0 = 0$
- **Symmetry**: any isometric transformation of coordinate space (t, X) that leaves **H invariant**, i.e. transforms solutions into other solutions
- **Allowed symmetry**: leaves $\Psi_A \Psi_B^*$, $\Psi_A \Psi_C^*$, ... (original beat wave) invariant
- **Allowed harmonics** (side bands): $\Psi_A^* d\Psi$, $\Psi_B^* d\Psi$, ... invariant for every allowed symmetry (multiply equation for $d\Psi$ with Ψ_A^* , Ψ_B^* , ...)
- **Even** symmetry ($\det=1$, translation, rotation, ...) determines position of harmonic peaks
- **Odd** symmetry ($\det=-1$, mirror symmetry, time inversion, ...) determines relations between **amplitudes** and **phases** of harmonic peaks, e.g. $a_+ = a_-$

Noether vs Laue: two sides of the same coin

Light ray reflecting off a grating. One direction is Noether, the other is Laue.

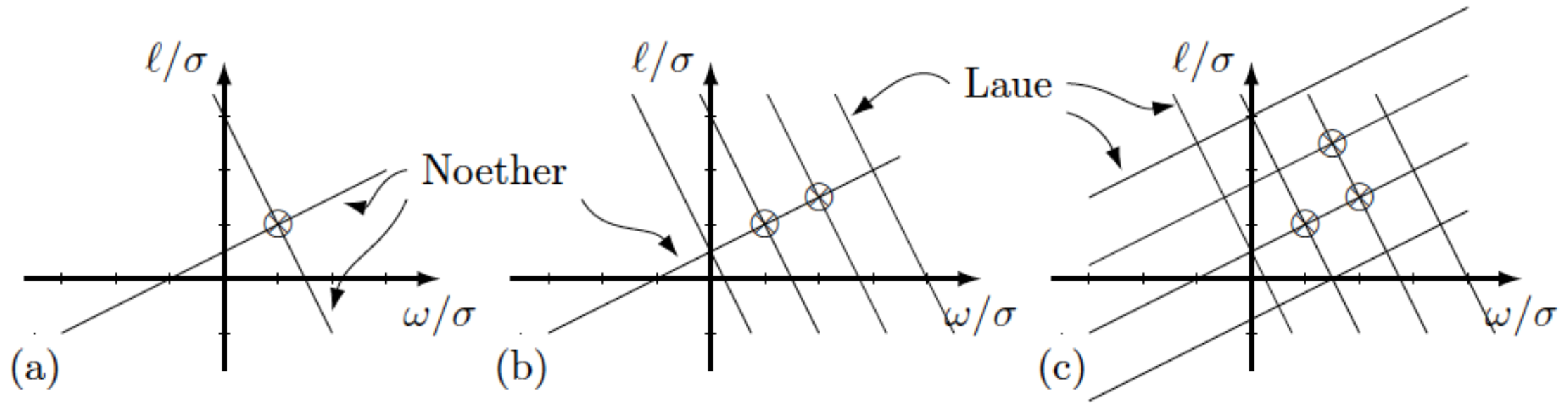


Friedrich W, Knipping P, Laue M, "Interferenz-Erscheinungen bei Röntgenstrahlen", Sitzungsberichte der Mathematisch-Physikalischen Klasse der Königlich-Bayerischen Akademie der Wissenschaften zu München, 1912: 303 (1912).

Noether, E., "Invariante Variationsprobleme". Nachrichten von der Gesellschaft der Wissenschaften zu Göttingen.

Mathematisch-Physikalische Klasse (1918): 235-257.

Symmetries vs harmonic progressions



- One fundamental mode: all (even) symmetries are continuous
- Noether's theorem: up to 4 Noether-conserved quantities
- Fundamental mode at intersection of these quantities
- No HHG progression

- Two fundamental modes: one discrete (broken) symmetry
- One (weakened) Noether quantity assumes a set of equidistant values
- Compare to Laue equations
- Harmonic modes at intersections
- 1-D HHG progression

- Three fundamental modes: two discrete (broken) symmetries
- Two Noether quantities each assume a set of equidistant values
- Harmonic modes at intersections of the Noether quantities (Laue again)
- 2-D HHG progression

This hierarchy of symmetries only becomes visible because we use “fundamental modes” with constant A^2 and ω/σ , k/σ etc., which most authors do not do

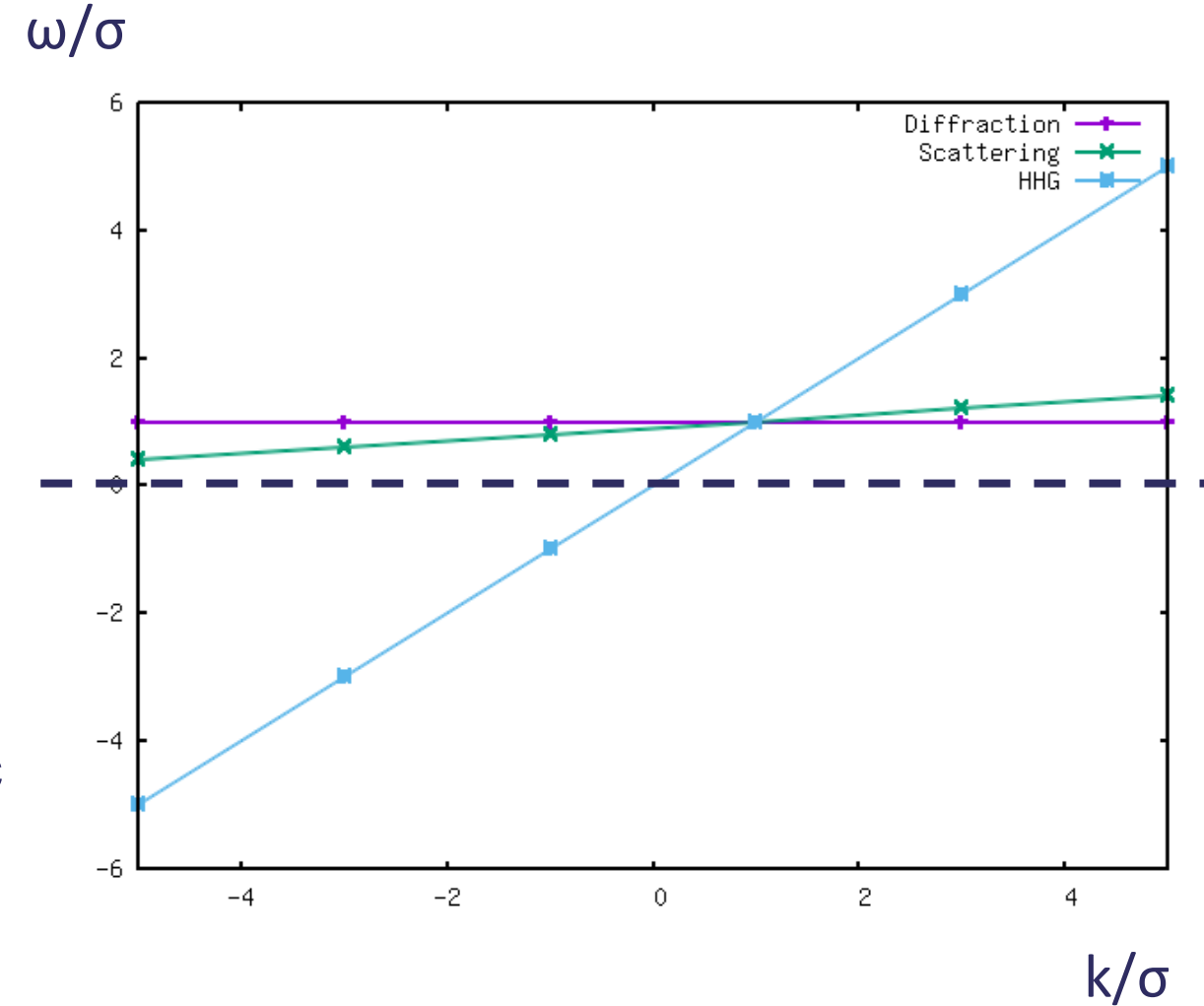
Connection with Bloch and Floquet theorems

	Our work	Mathieu	Floquet	Bloch
Starting point (translation e.f.)	$\Psi_{A,B} = \exp(i \cdot K_{A,B} \cdot X)$	$\exp(\pm i \cdot x \sqrt{a})$	$\exp(t \cdot B)$, B a square matrix	Plane wave $\exp(i \cdot \mathbf{k} \cdot \mathbf{x})$
These terms determines the allowed symmetries	$\Psi_A \Psi_B^*$, $\Psi_A \Psi_C^*$; these are periodic	$2q \cdot \cos(2x)$	Matrix A with $A(t+T) = A(t)$	Periodic (lattice) potential $V(x)$
This rule sets the harmonics Ψ_n wrt starting point	$\Psi_A^* \Psi_n$, $\Psi_B^* \Psi_n$ invariant under symmetries	$\Psi_n \exp(\mp i \cdot x \sqrt{a})$ invariant under symmetries	$\Psi(t) \cdot \exp(-t \cdot B) = P(t)$, $P(t+T) = P(t)$	$\Psi(t) \cdot \exp(-i \cdot \mathbf{k} \cdot \mathbf{x})$ periodic like $V(x)$

Variations on a theme!

A final thought...

- $\Delta\omega = 0$: diffraction
 - $0 < \Delta\omega < \omega_0$: scattering (e.g. Raman, Brillouin)
 - $\Delta\omega > \omega_0$: harmonic generation
 - But how different are these processes really?
-
- Diffraction: stationary grating
 - Scattering: slowly moving grating
 - Harmonic generation: grating moves at c and spacing less than λ_0
 - The same idea every time...



Conclusions

We have developed **a novel way** to describe, study and diagnose harmonics

- Decompose all EM waves into fundamental CP modes; DC modes to describe targets
- All $(\omega/\sigma, \mathbf{k}/\sigma)$ spectra look like **regular grids**; explains many **existing results**
- We can generate spectra **to order** using structured targets

Frequency combs

- Every **2-D progression** has the potential for a **frequency comb**
- Use **two LP pulses** with different **OAM levels** or **transverse angles**, or one pulse with a corrugated target, to create a 2-D progression

Symmetry theory

- Harmonic spectra can also be described via **symmetry theory**, symmetries of Ψ_0^2
- Direct links to **Noether's Theorem** (cont. symm.) and the **Laue equations** (discrete symm.)
- Further connections to **Mathieu**, **Floquet** and **Bloch** theory

Beyond plasma physics: HHG in **chiral media**

- See R. Trines et al., arXiv:2604.08354 (2026).

R. Trines et al., Nature Comms. **15**, 6878 (2024).

R. Trines et al., Phys. Rev. Research **8**, 013241 (2026).

R. Trines et al., arXiv:2507.08635 (2025).

R. Trines et al., arXiv:2604.08354 (2026).

Bonus: HHG in chiral media

- We have expanded our beat-wave approach to “**asymmetrical**” HHG in **asymmetrical** (chiral) molecules (molecule has **left-** and **right-**handed versions)
- Not enough time now, but just to whet your appetite
- Interaction of laser light with the “**transverse spin**” of a **focused** beam leads to a (bonus) **chiral DC mode**, which changes **sign** when you change the **handedness** of your molecule
- This DC mode, together with the (achiral) laser modes, generates a harmonic “**chessboard**” spectrum where half the modes are **chiral** and half achiral
- Thus, if you change the handedness of your molecule, **half** your harmonics will **change sign** and half will not. This can be analysed in many ways

In our paper, we derive:

- The **chiral DC** mode for suitable types of structured light
- **Rules** that distinguish between various **types of chirality** (mostly global vs local)
- **Rules to switch** from one type of chirality to another



Science and
Technology
Facilities Council

Thank you

Facebook: Science and
Technology Facilities Council

Twitter: @STFC_matters

YouTube: Science and
Technology Facilities Council