seit 1558

Strong-field physics meets quantum optics

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NOA NONLINEAR OPTICS DOWN TO ATOMIC SCALES

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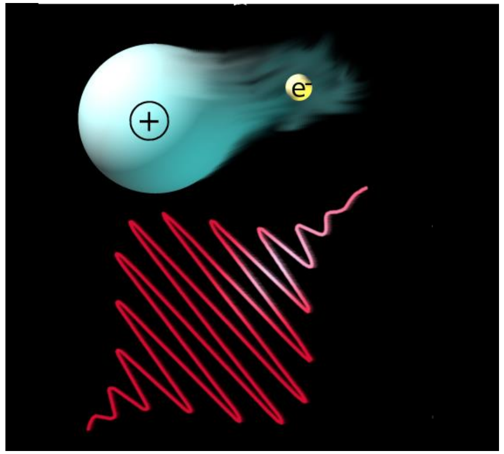
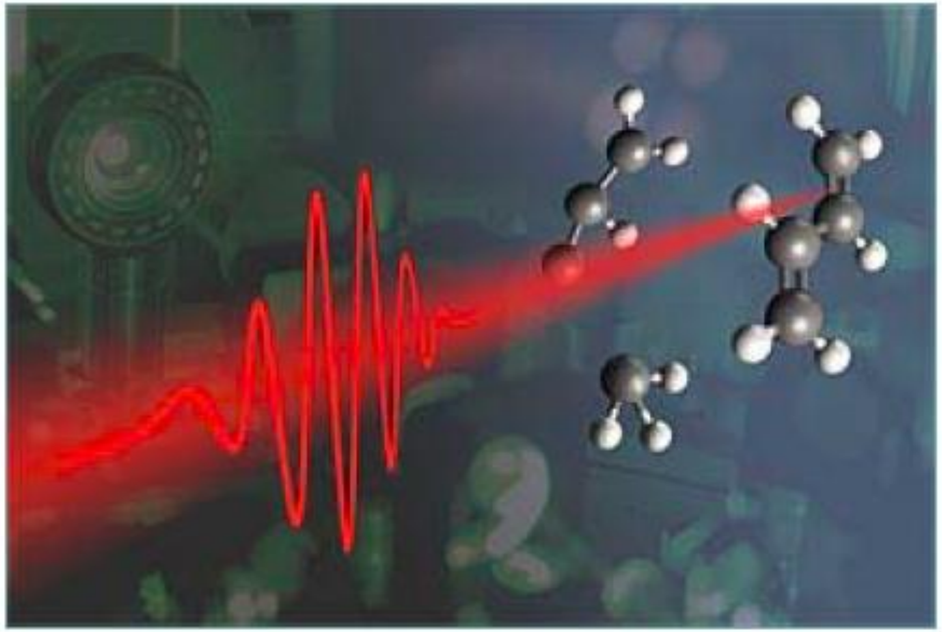
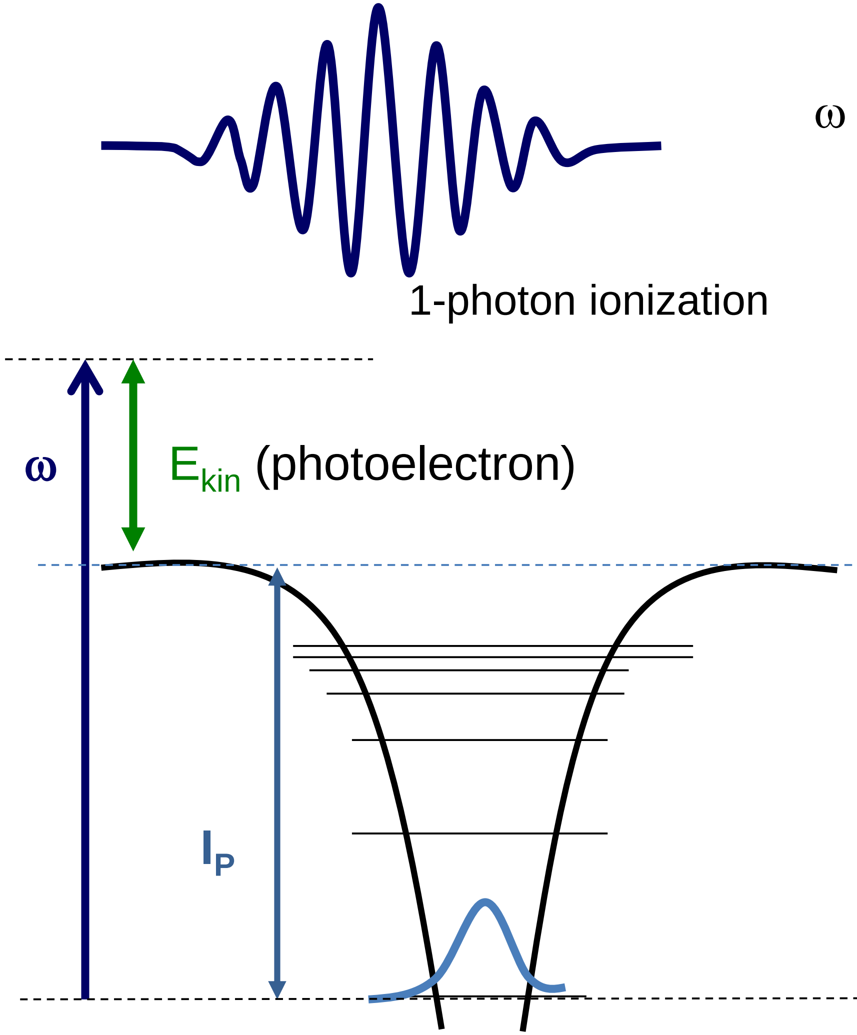




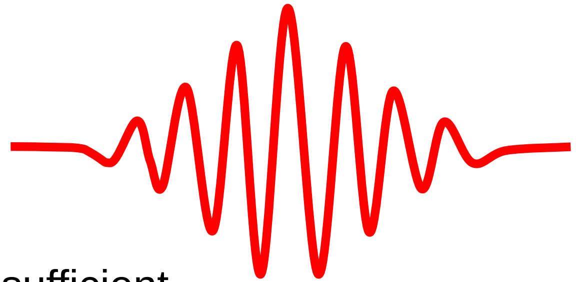
Basic introduction: Strong-field light-matter interaction

Ionization

Ionization:
$\omega > I_P$ vs. $\omega < I_P$



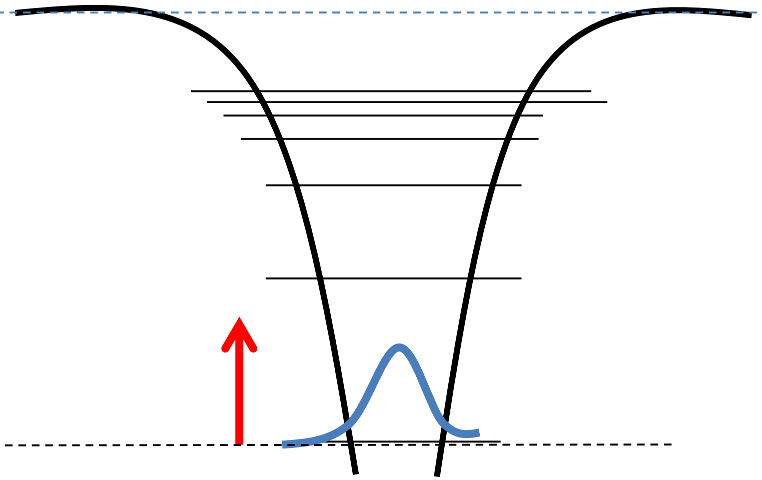
Perturbative treatment



1 photon insufficient
for ionization

→ more than one photon

→ higher intensities



Perturbative treatment not possible

Strong-field ionization (low frequency limit)

Keldysh parameter

$$\gamma \equiv \sqrt{\frac{I_P}{2U_P}} = \omega\tau$$

$$= \frac{\tau_{tunnel}}{\tau_{optical}}$$

$$\tau = \frac{v_{el}}{E_0} = \frac{\sqrt{2I_P}}{E_0}$$ „tunneling time“

$$U_P = \frac{(E_0)^2}{4\omega^2}$$

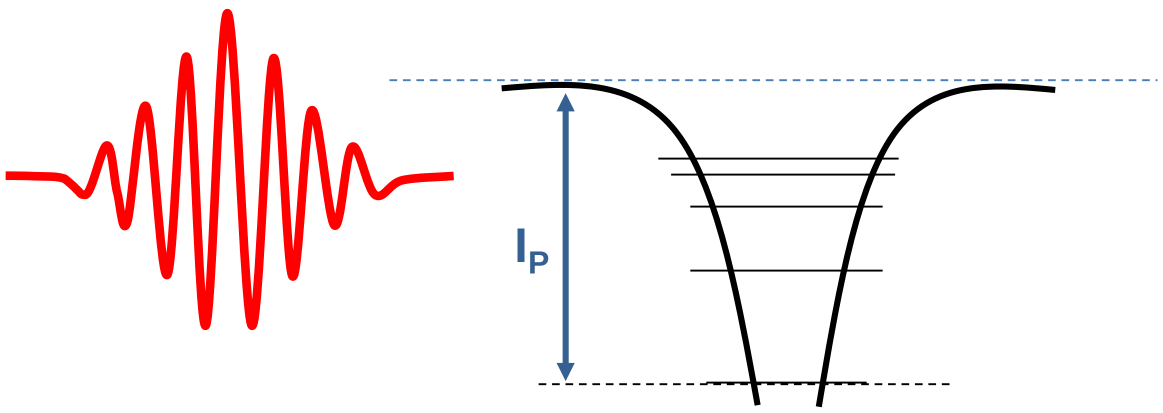
Ponderomotive potential
(„kin. energy of e^- in laser field“)

“slow” → tunneling

$$\gamma << 1$$

“fast” → multiphoton

$$\gamma >> 1$$



M. Yu. Ivanov, M. Spanner, O. Smirnova, J. Mod. Optics 20, 165 (2005)
“Anatomy of strong-field ionization”

Strong-field ionization: mechanisms

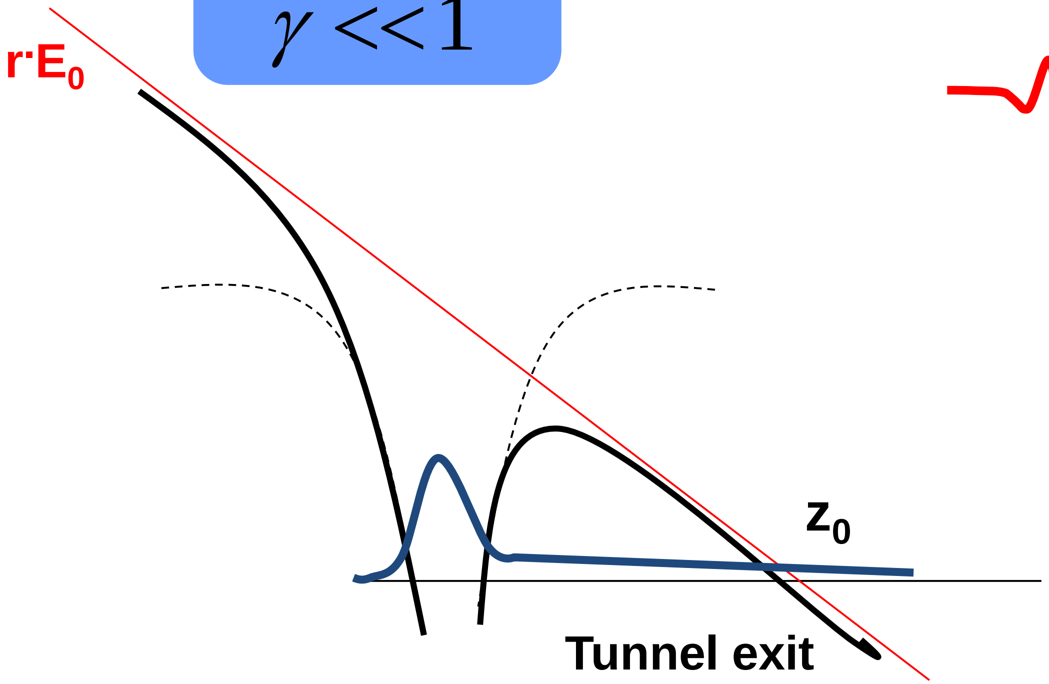
Keldysh parameter

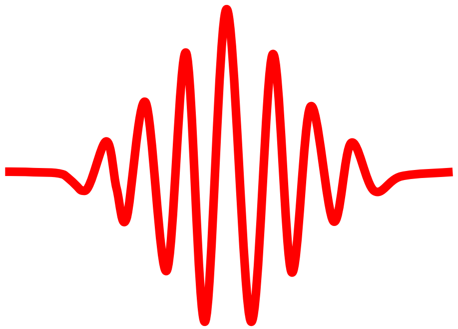
$$\gamma = \sqrt{\frac{I_P}{2U_P}}$$

$$U_P = \frac{(E_0)^2}{4\omega^2}$$ Ponderomotive potential

Tunnel regime

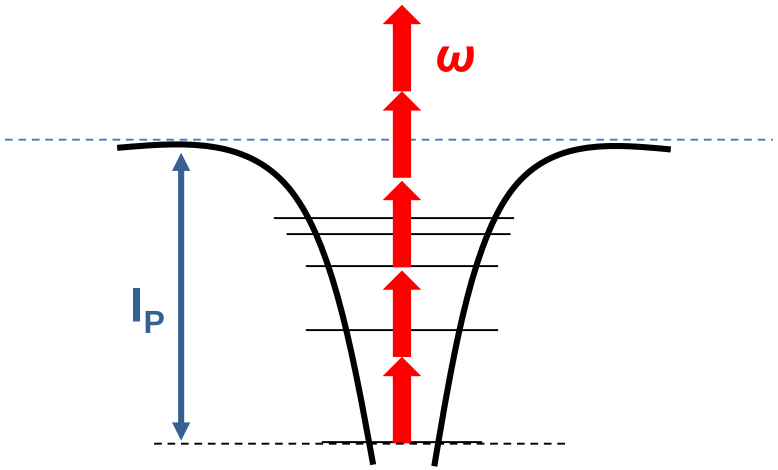
$$\gamma \ll 1$$





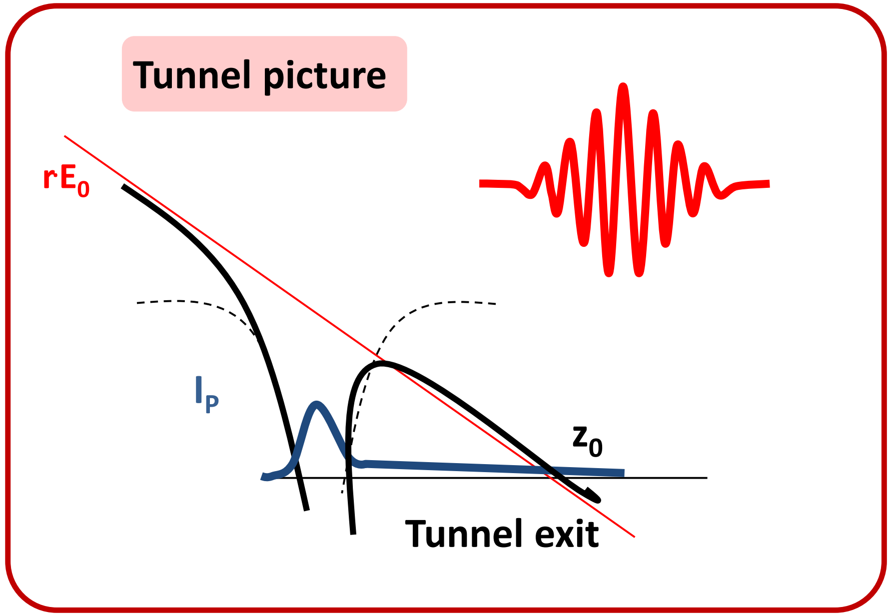
Multiphoton regime

$$\gamma \gg 1$$



M. Yu. Ivanov, M. Spanner, O. Smirnova, J. Mod. Optics 20, 165 (2005) "Anatomy of strong-field ionization"

Introduction: Strong-field physics



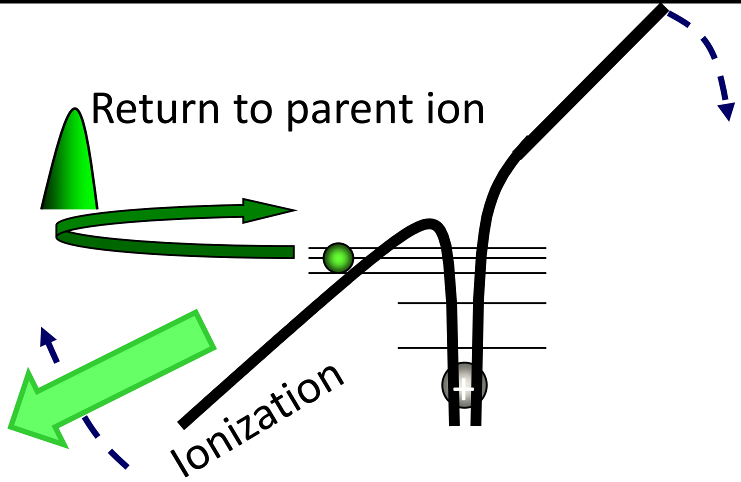
$$U_P = \frac{E^2}{4\omega^2}$$

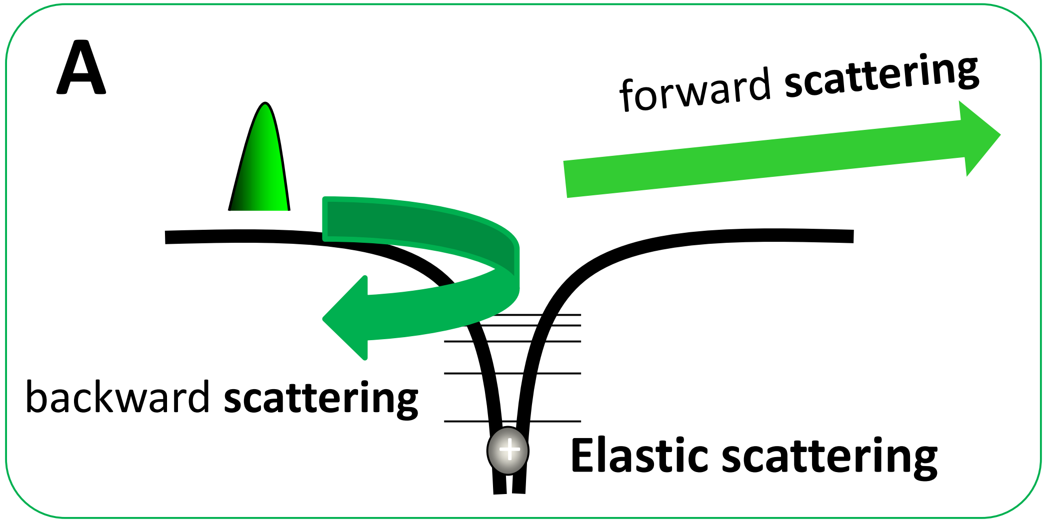
Av. kin. energy of e⁻ in the laser field

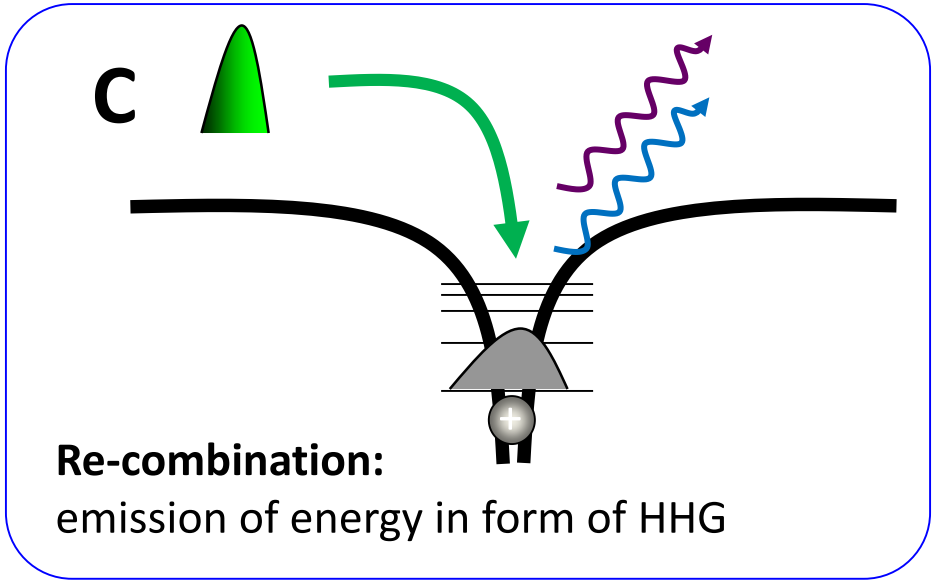
$$\tilde{\alpha} = \frac{E}{\omega^2}$$

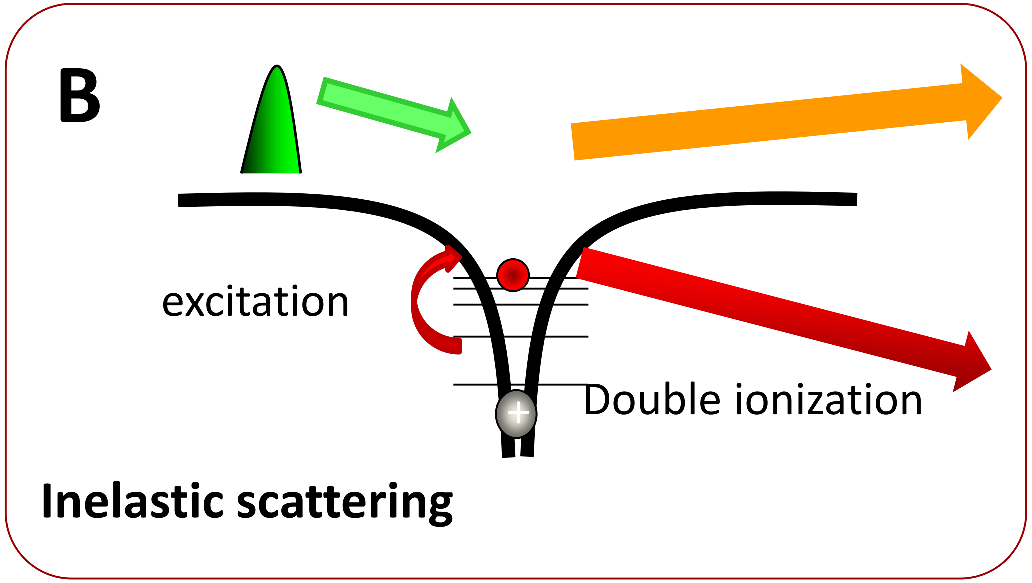
Quiver motion of e^- in the laser field

What happens after ionization?
Electron dynamics in intense laser fields









High-order harmonic generation

Lower-order (perturbative) harmonics decrease in intensity; high-order harmonics form a plateau

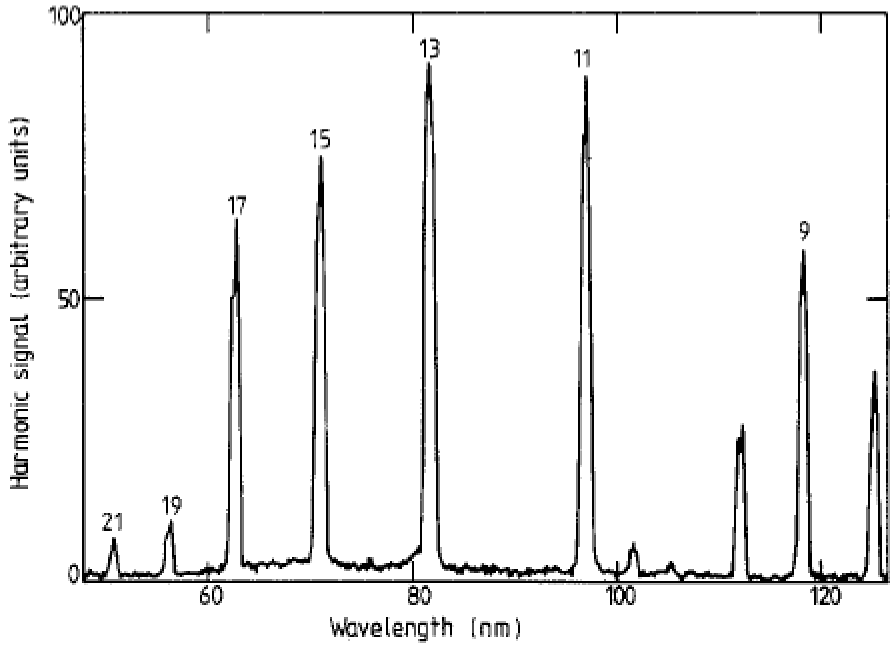
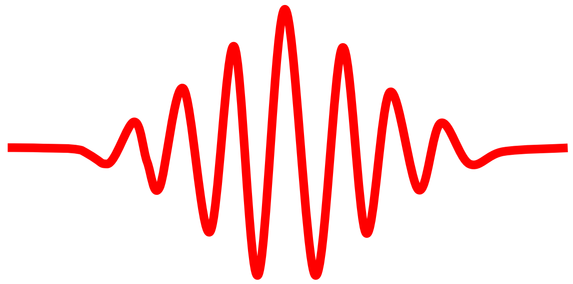


Figure 1. Harmonic spectrum obtained using a Xe gas jet showing all odd harmonics between 9 and 21. The peaks at 101, 112 and 125 nm are the second diffracted orders of the 21st, 19th and 17th harmonics respectively. The laser intensity was approximately 3×10^{13} W cm^{-2} and the Xe pressure at the focal point was about 10 Torr.



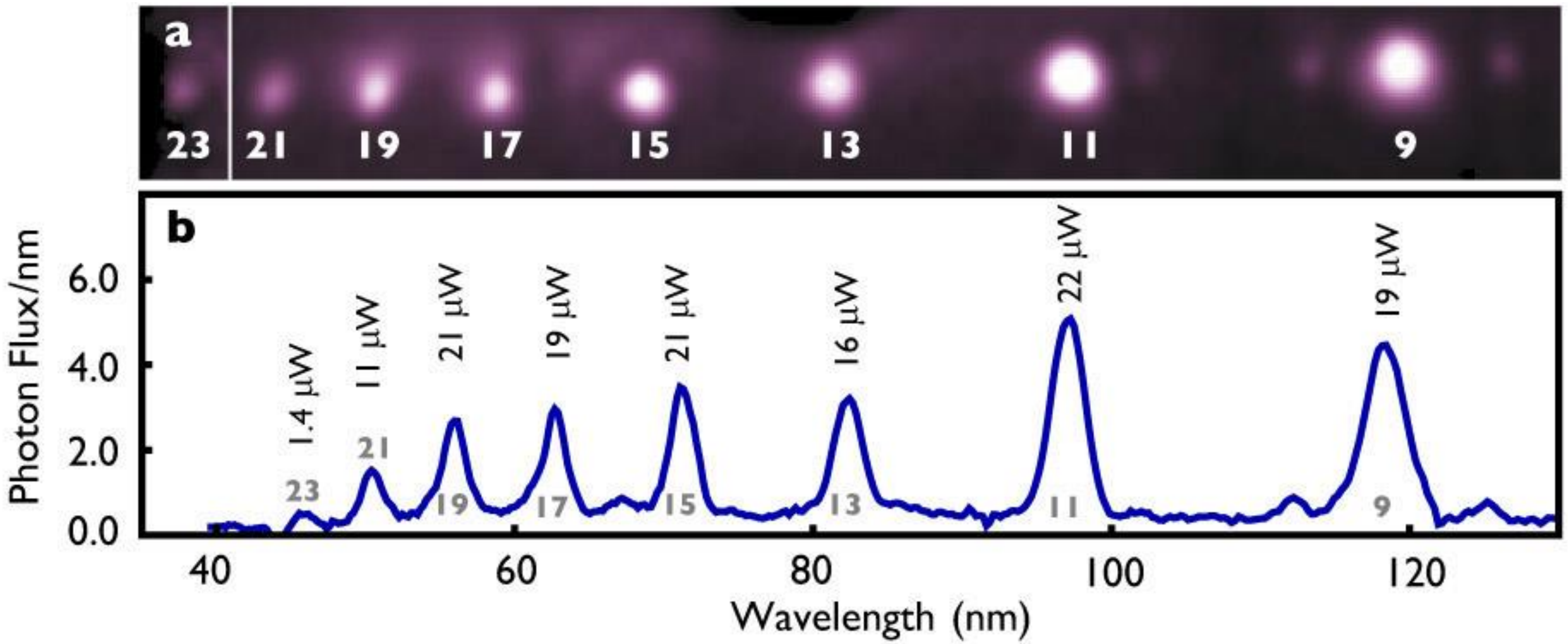
laser parameter:
1064 nm, ~30 ps, 3×10^{13} W/cm^2

M. Ferray, A L'Huillier, XF Li, LA Lompré, G Mainfray, C Manus, "Multiple-harmonic conversion of 1064 nm radiation in rare gases", J Phys B. 21, L31 (1988)

Only odd harmonics

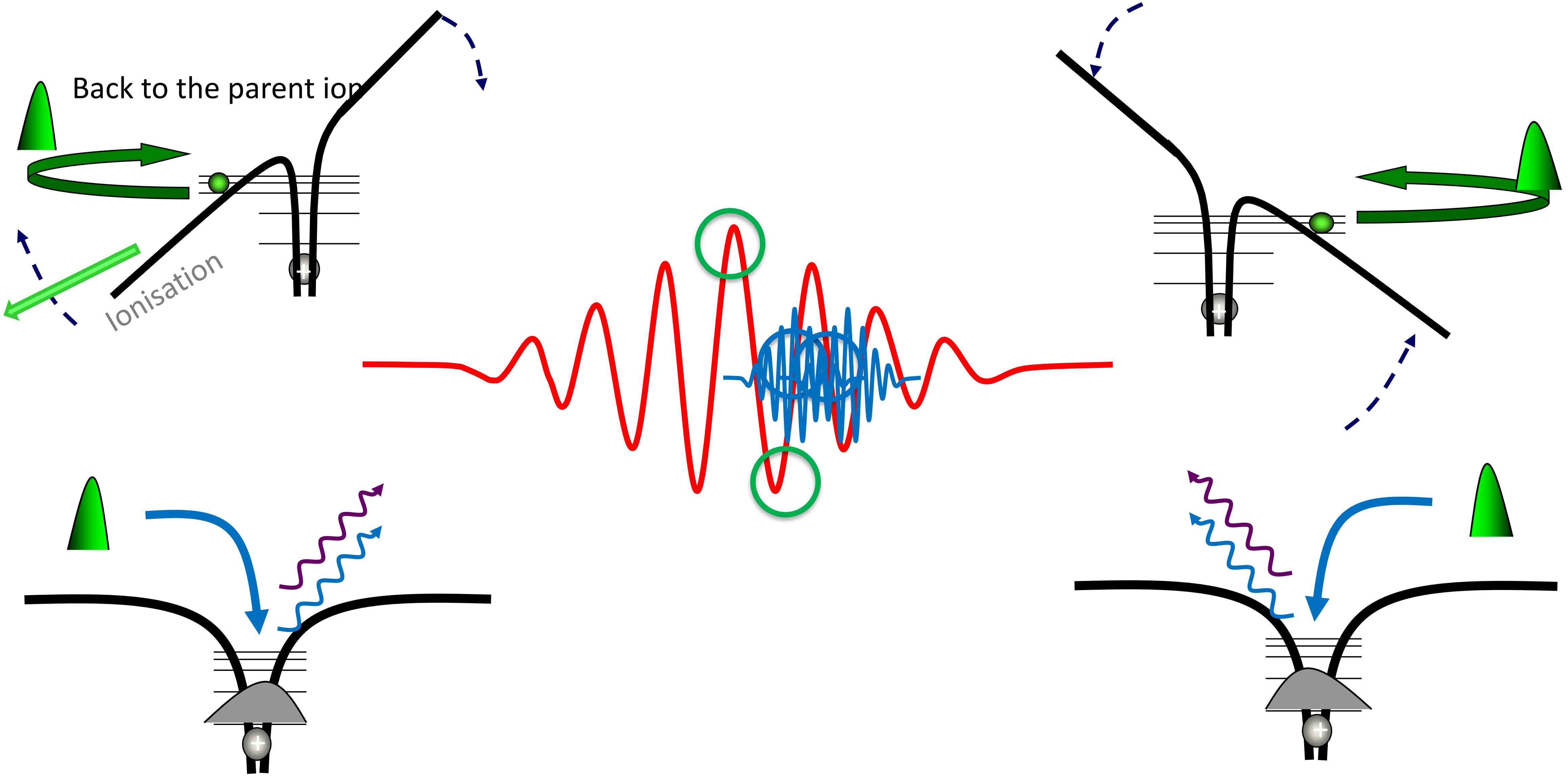
Why are in an HHG spectrum
(a) Peaks?
(b) only odd-order peaks?

<u>Peak structure in HHG spectra:</u>
Fourier-relation: regular pattern in frequency domain – must be regular process in time domain

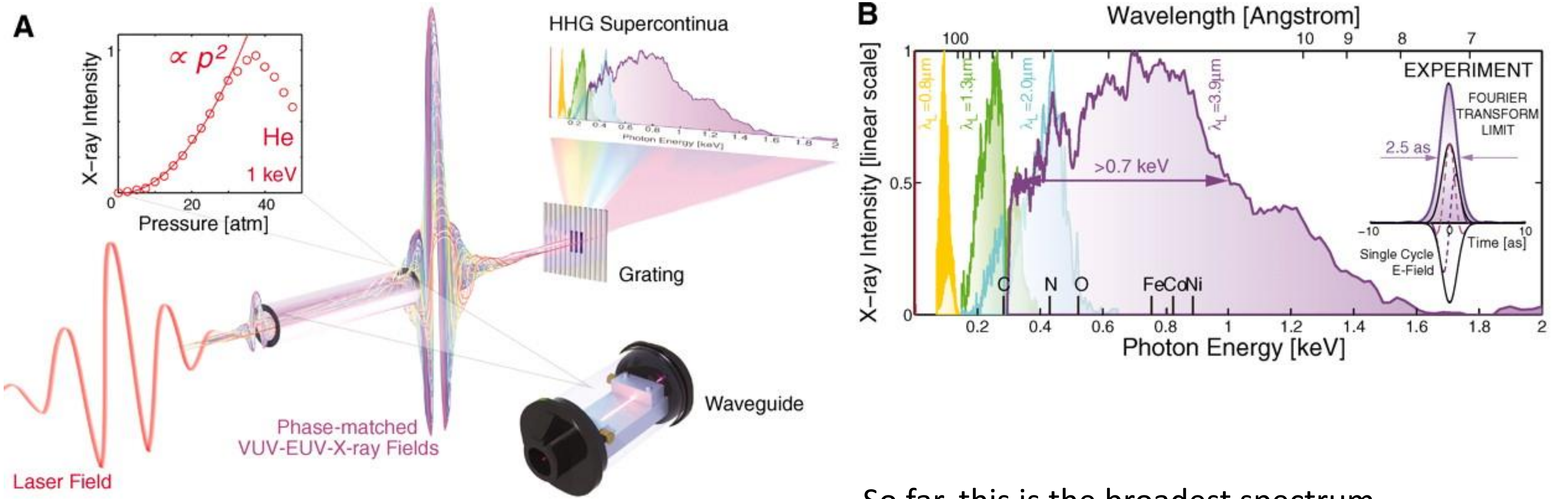


Taken from: A Cingöz, DC Yost, TK Allison, A Ruehl, ME Fermann, I Hartl, J Ye,
"Direct frequency comb spectroscopy in the extreme ultraviolet" Nature 482, 68 (2012)

Repeating process : every half-cycle (2x per optical cycle)



The „shortest” pulse!?

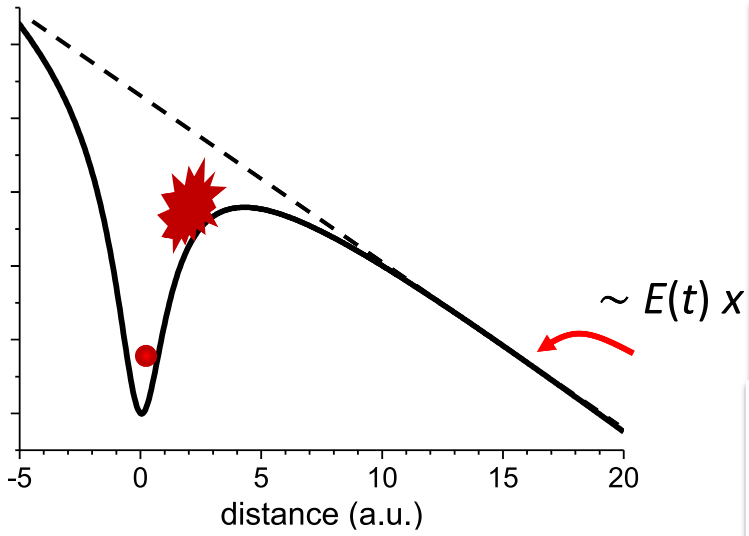


So far, this is the broadest spectrum
which would correspond to the shortest pulse;
however, no way for direct measurement

T Popmintchev, et al., MM Murnane, HC Kapteyn, “Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers”, Science 336, 1287 (2012)

High Harmonics Generation (HHG) – Gas vs. condensed phase

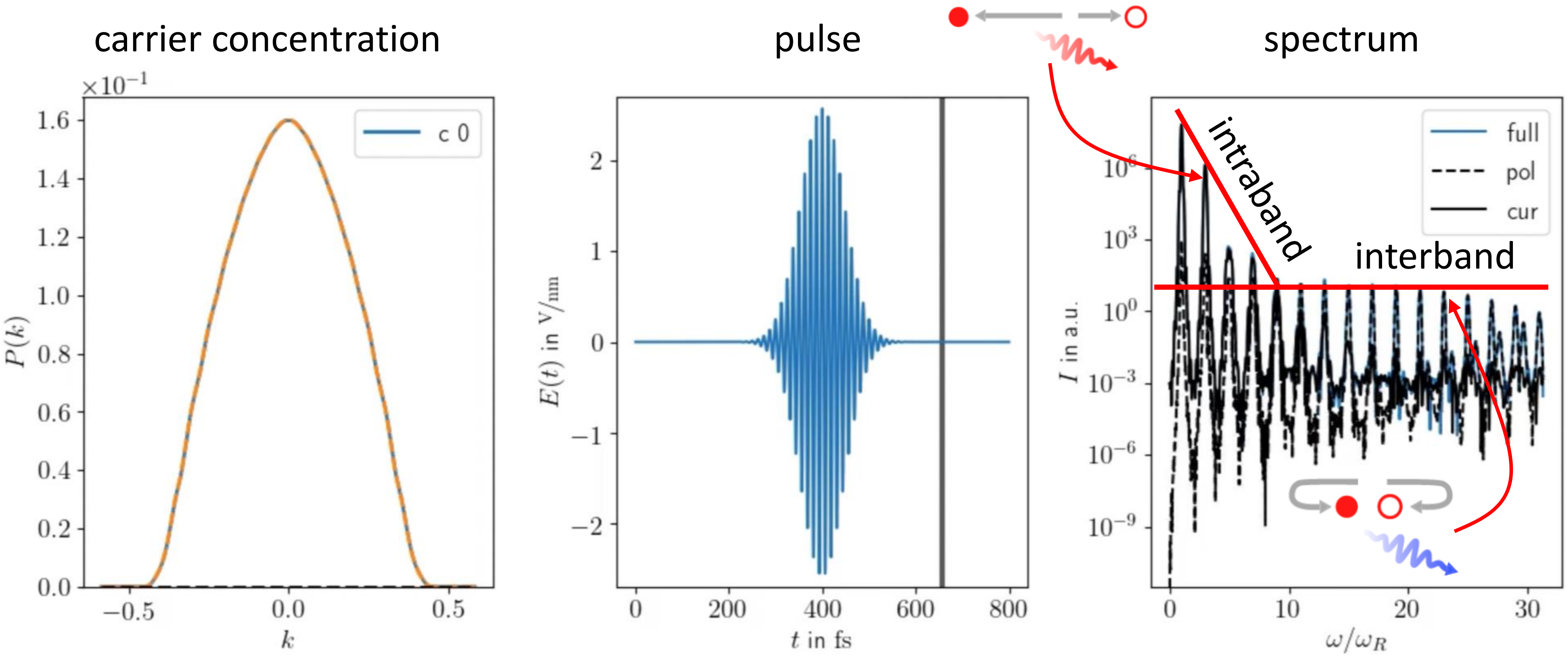
Atomic/molecular gases



Dielectrics /semiconductors

Figure: Taken from DOI:

High Harmonics Generation (HHG) in dielectrics - simulation

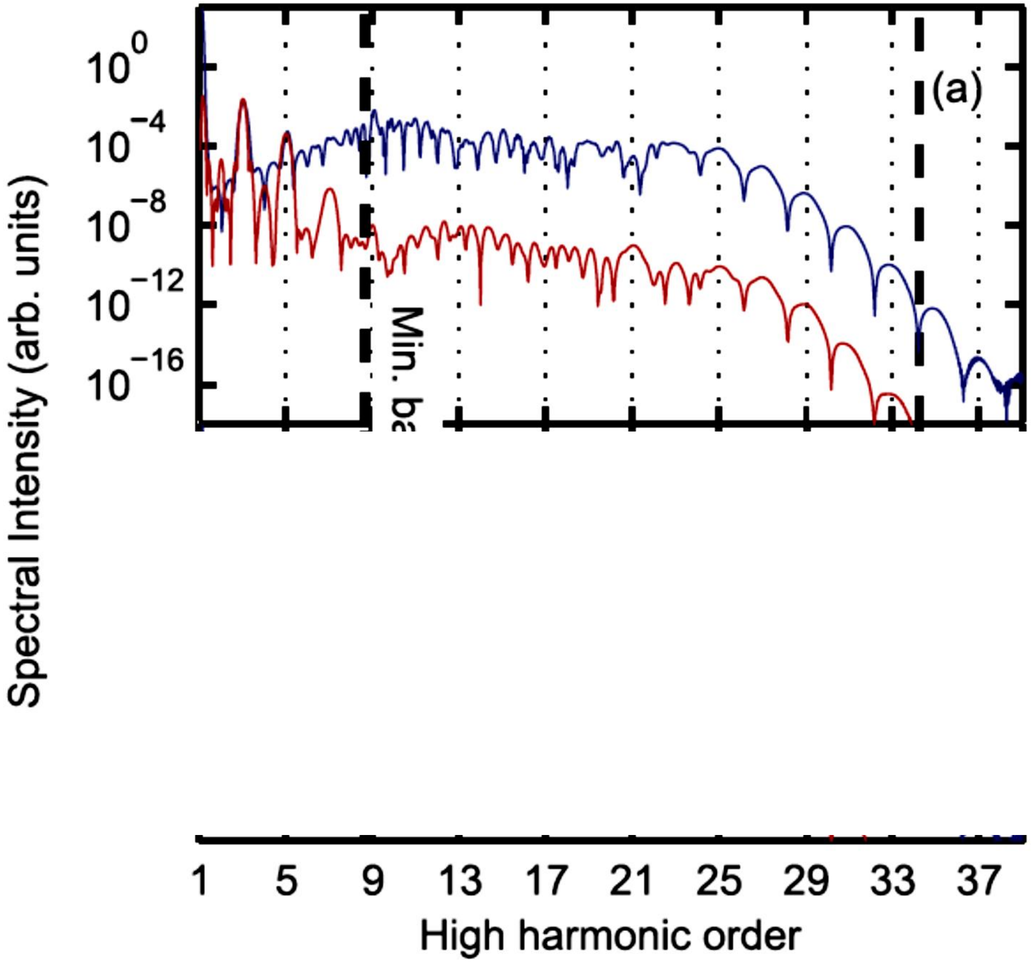


Simulations in momentum space based on Semiconductor Maxwell-Bloch Equations

Material: ZnO, gap energy 3.3eV (375nm) pulse: 100fs λ=4μm E_{max}= 2.3 V/nm

D. Golde, T. Meier, and S. W. Koch, High harmonics generated in semiconductor nanostructures by the coupled dynamics of optical inter- and intraband excitations, Phys. Rev. B 77, 075330 (2008).

The dephasing time



No dephasing time: no distinct harmonics

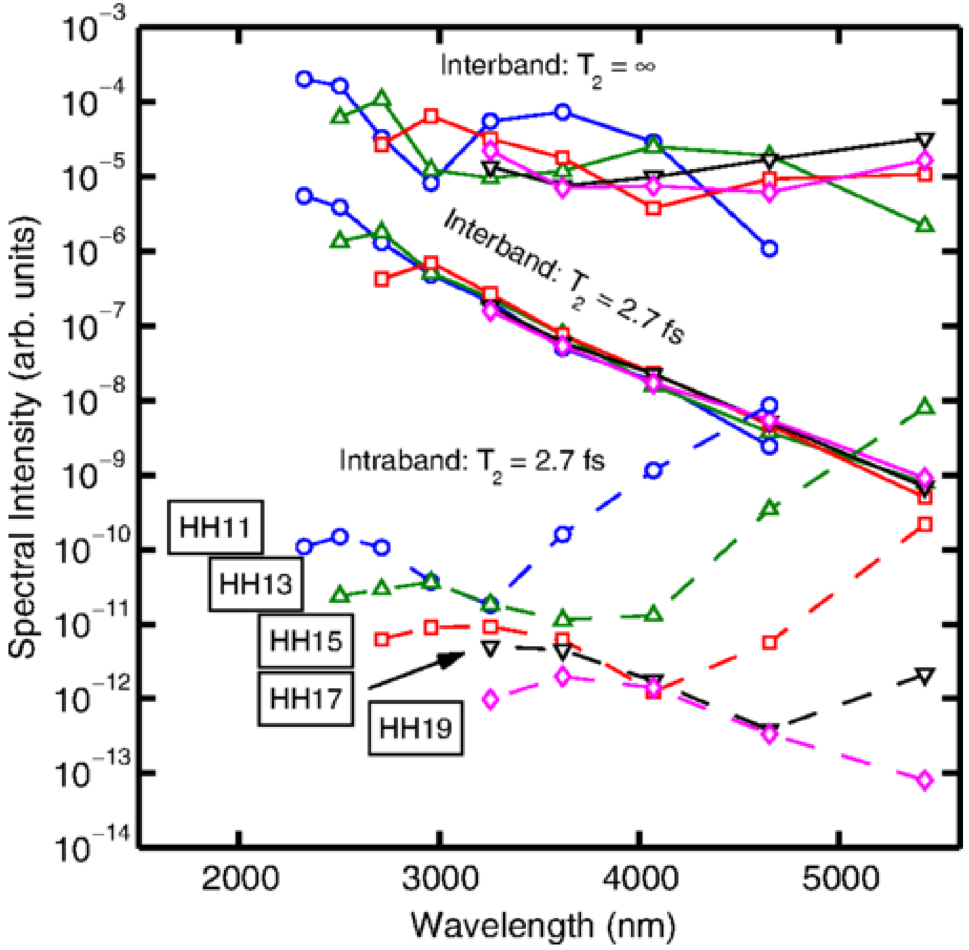
Introduction of dephasing times:
T_1: decay time of carrier population;
T_2: decay time of the polarization (decay of quantum coherence in coherently excited system)

Phenomenological values for T_2 ~ 2 fs – less than a 1/4 of optical cycle (??)
Necessary to match simulated and experimental spectra

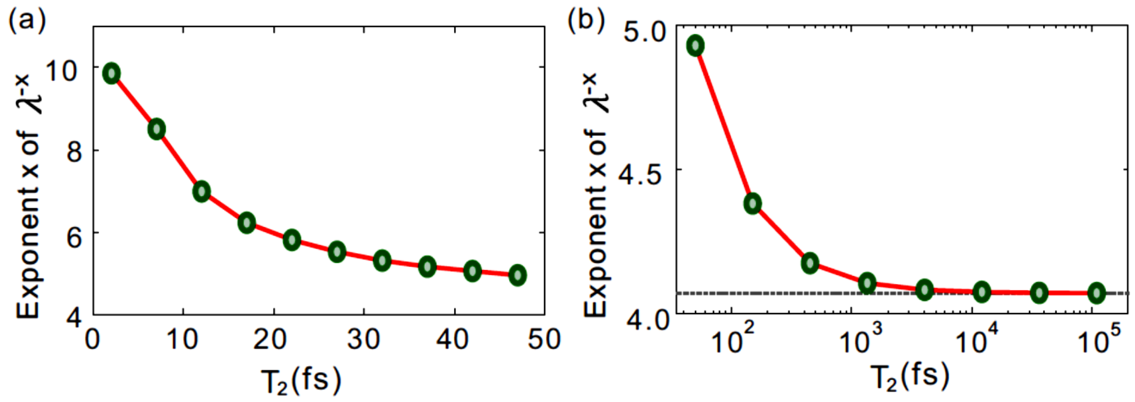
Vampa et al., PRL 113, 073901 *(2014)*

The „dephasing time“

Dephasing defines the exponential decay of harmonic yield with driving wavelength



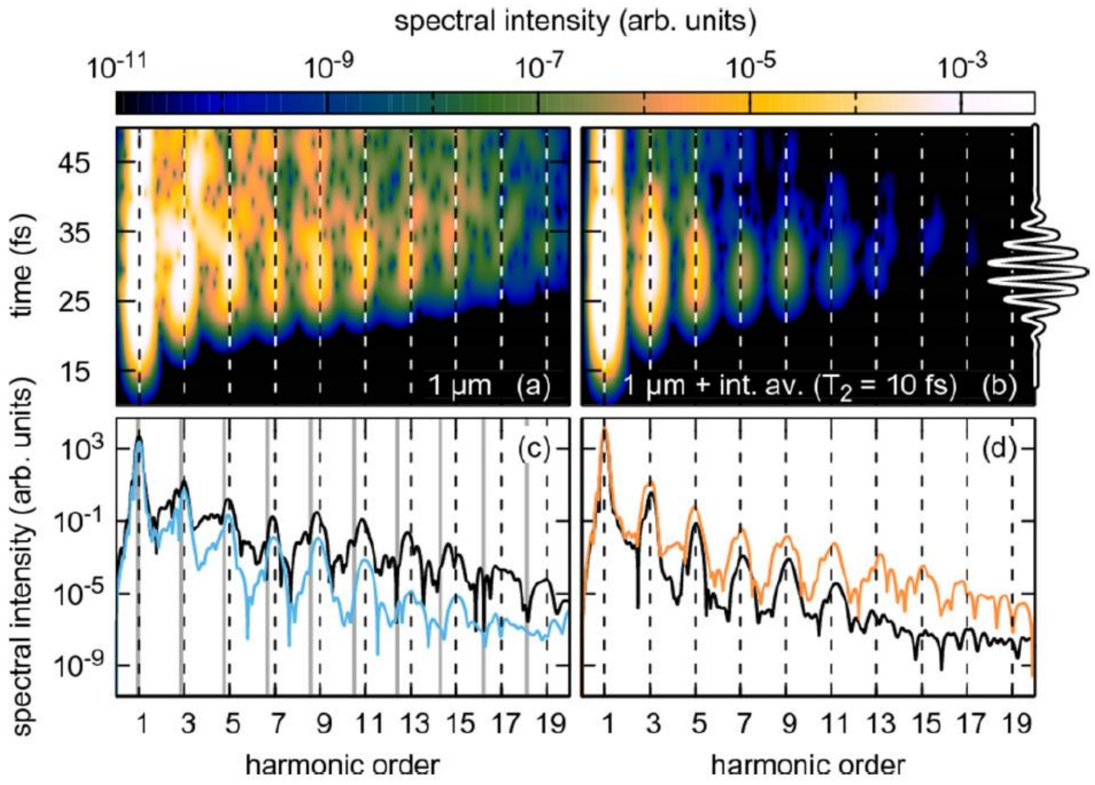
Vampa et al., PRL 113, 073901 *(2014)*



X. Liu, …. Peixan Lu, PRA 98, 063419 (2018)

- ➢ **What is the wavelength dependence of HHG in solids?**
- ➢ **What is the origin of the dephasing?**

Ultrafast dephasing time T_2?



What is the physical origin of such ultrafast dephasing?

Hypothesis: **propagation effects**: "Ultrafast microscopic dephasing rates of the order of $T_2 \approx 1$ fs previously invoked are neither necessary nor justified for forming a well-defined harmonic spectrum. "

->No dephasing or at least order of magnitude larger dephasing time + propagation effects!

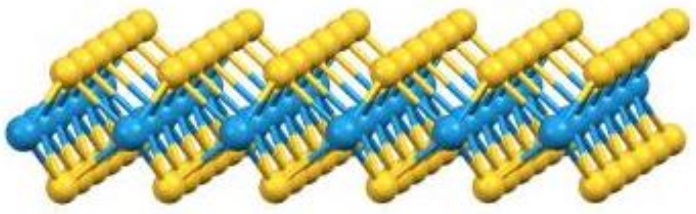
I. Floss, …. Yabana, Burgdöfer, PRA 97, 011401(R) (2018)

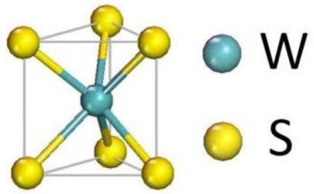
-- Let's investigate

Wavelength-dependence of HHG in thin samples

Samples

- **Single atomic layer WS_2 or MoS_2**
- **140 nm thick wurtzite polycrystalline CdSe film**



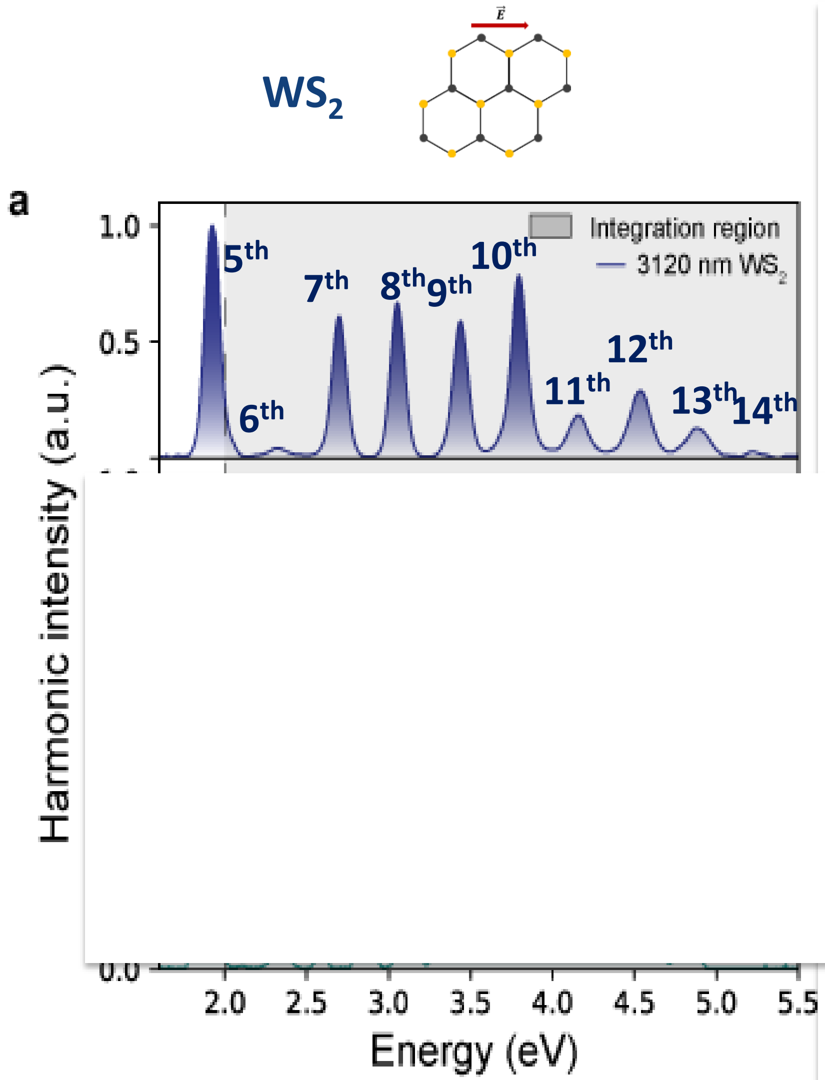


No propagation effects!

Experiments by Daniil Kartashov

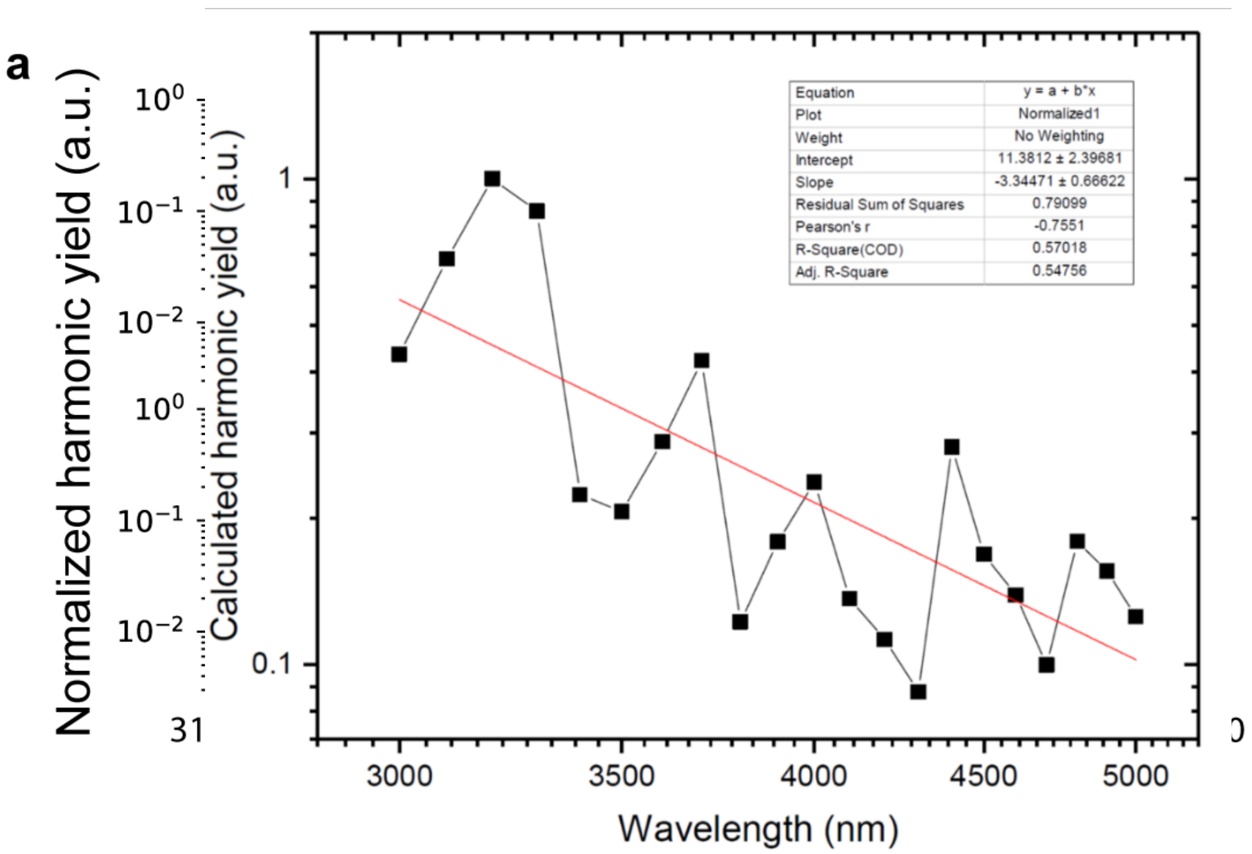
V. Korolev et al., „Unveiling the Role of Electron-Phonon Scattering in Dephasing High-Order Harmonics in Solids", submitted**; arXiv:2401.12929**

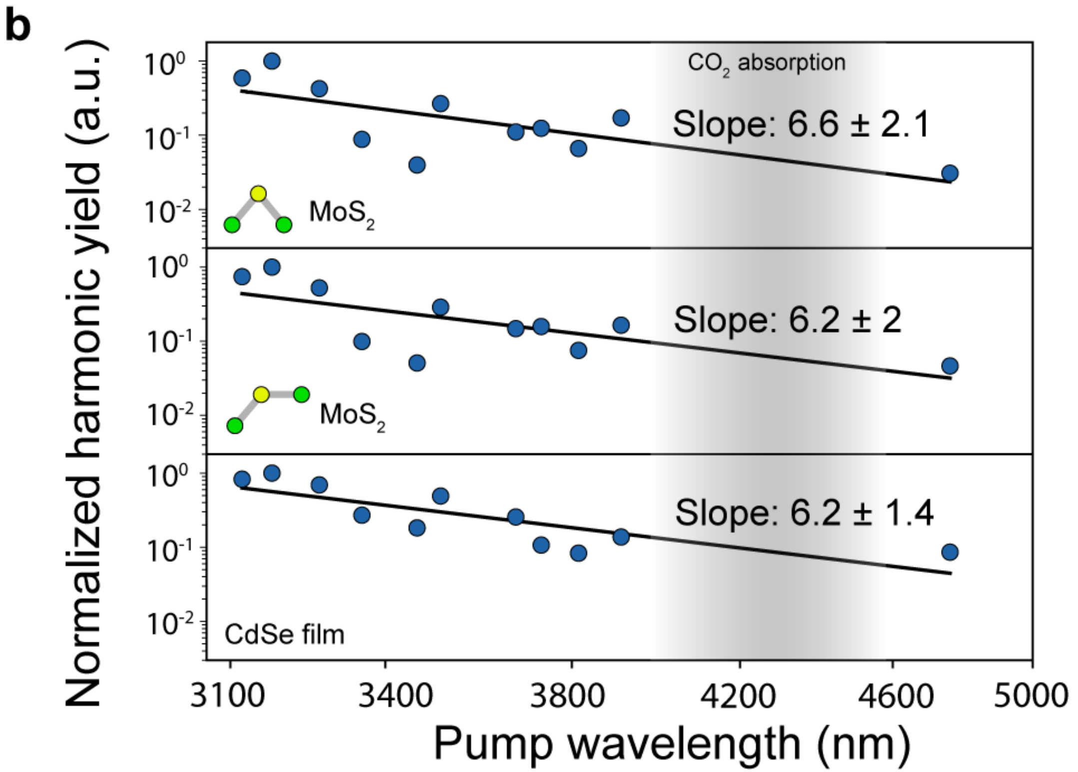
Experimental results



Integral harmonic yield in the range 2 (1.9) – 5.5 eV

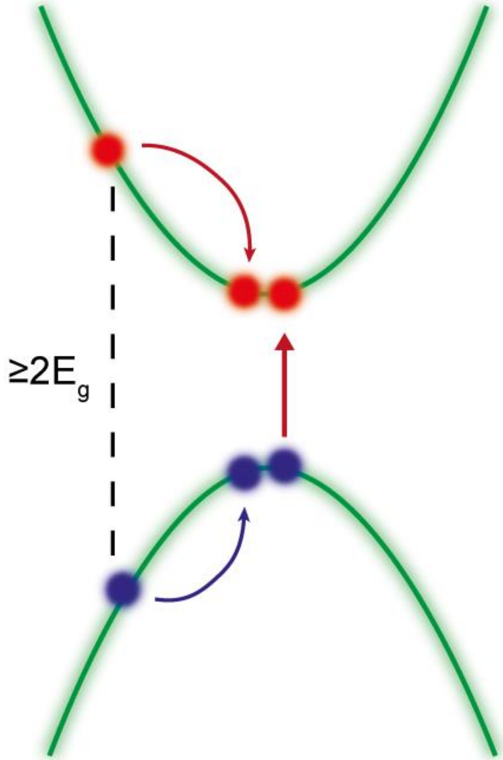
Wavelength dependence in HHG

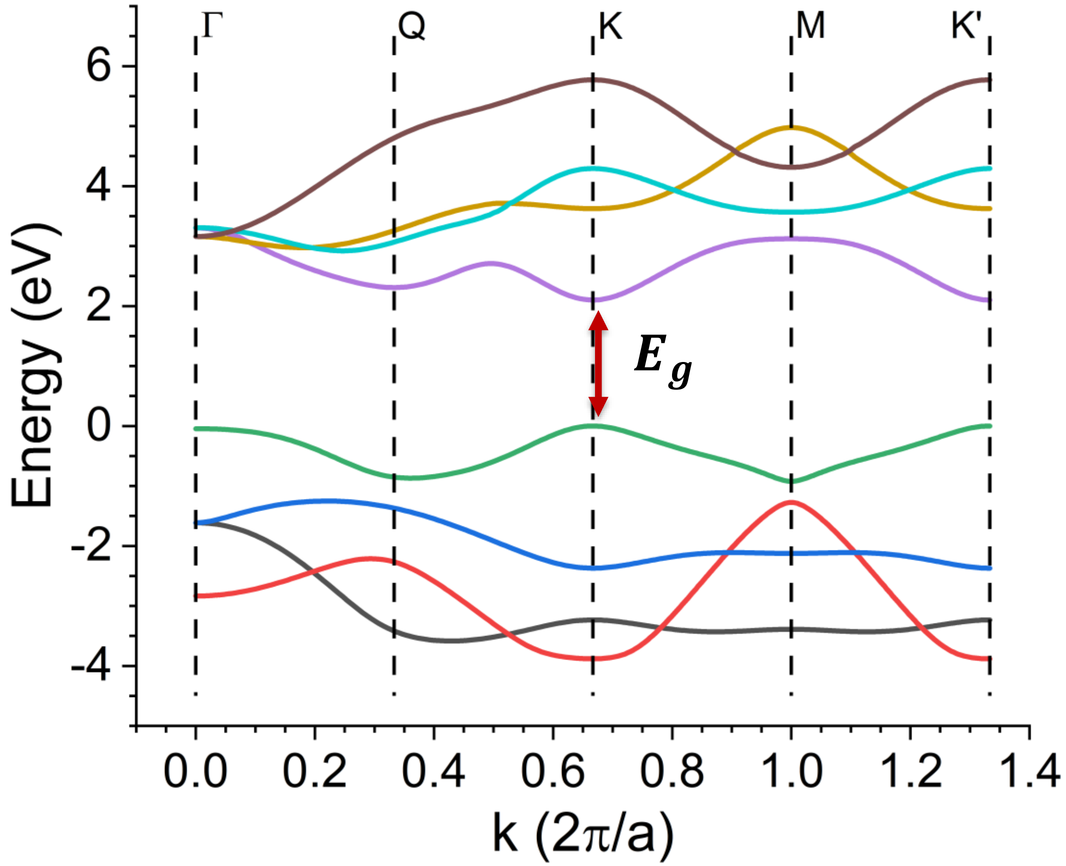




Numerical simulations without dephasing predict $\lambda^{-3.3}$ (rt-TDDFT) or λ^{-4} (SBE) dependence!

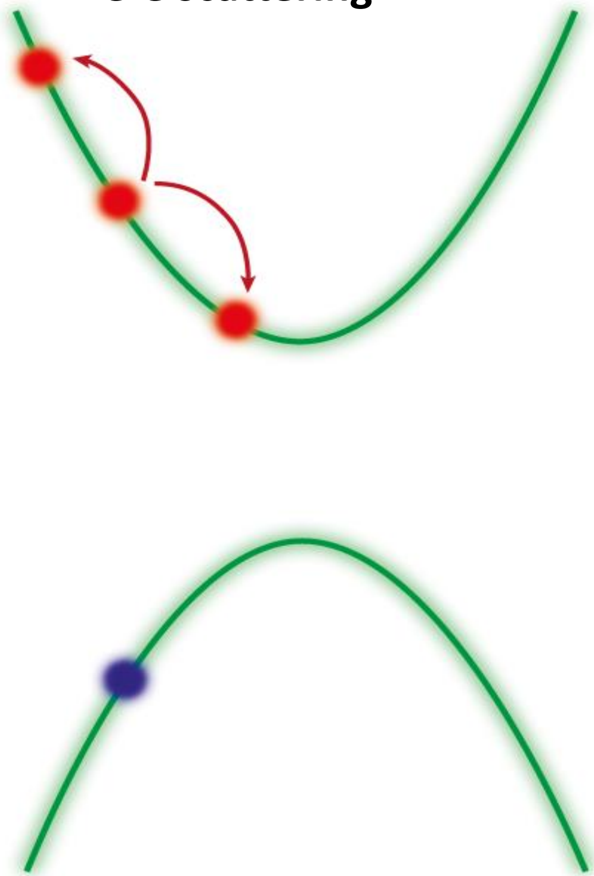
Origin of the dephasing: Carrier multiplication?

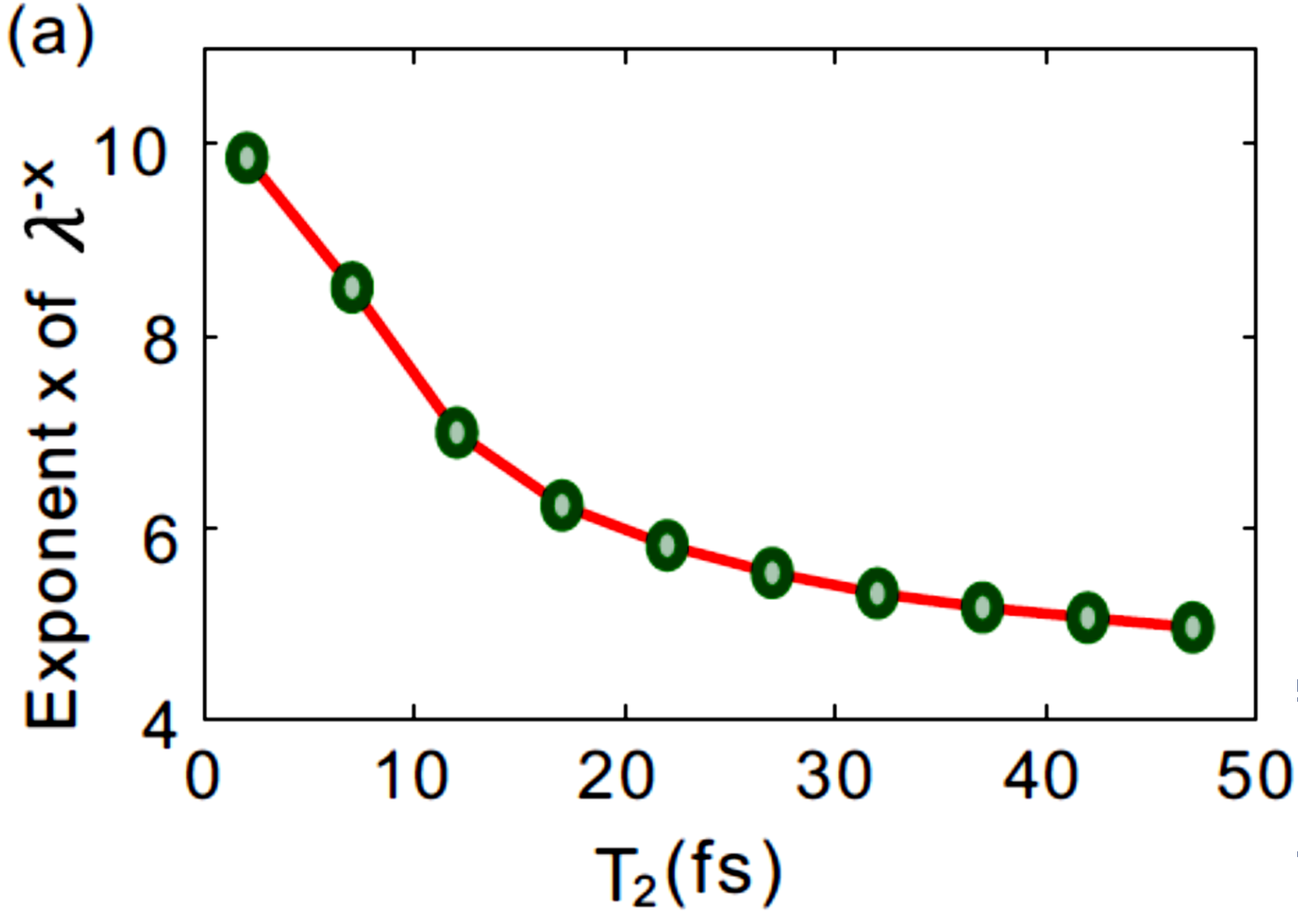




Requires kinetic energy >2 E_g - inefficient for electron motion within the band

Origin of the dephasing: e^- -e^- scattering?





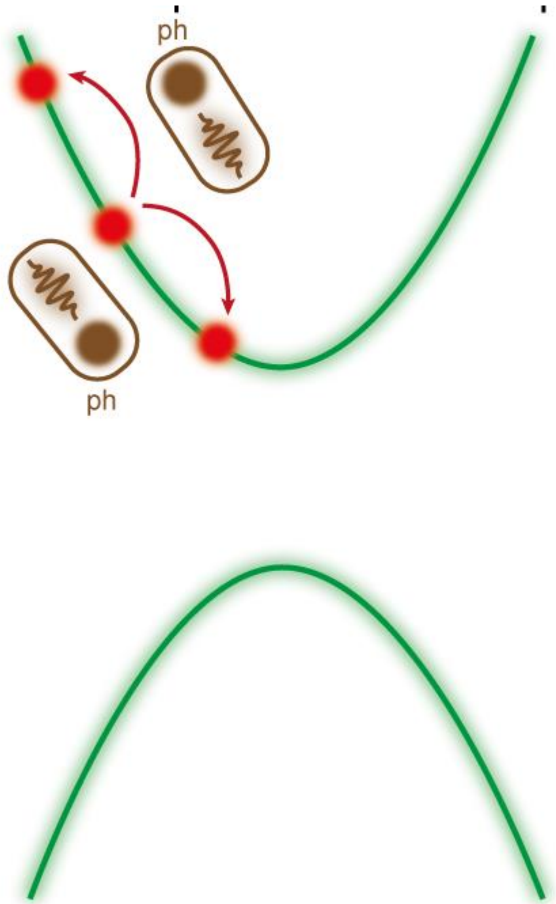
estimated

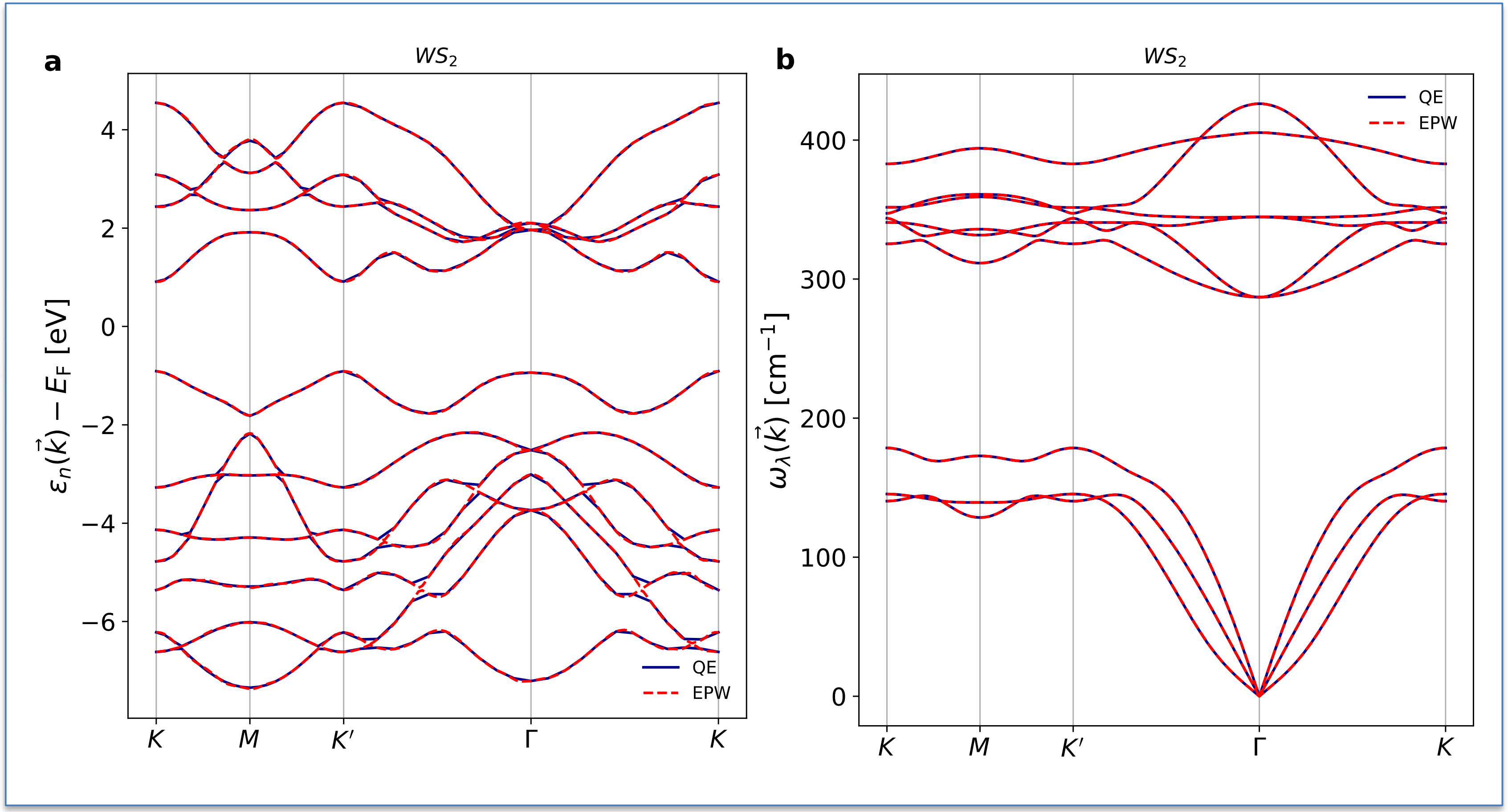
he wavelength

dependence of the yield

Liu et al., PRA 2018

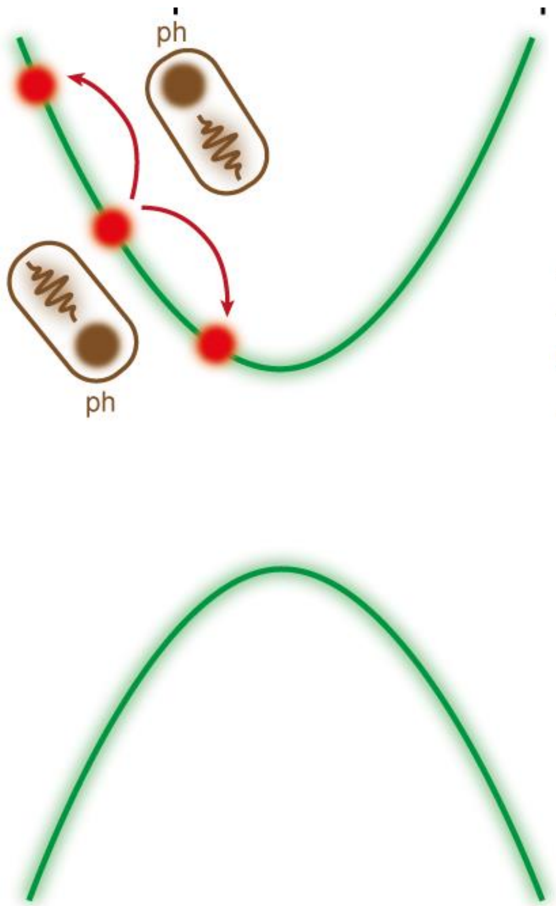
Origin of the dephasing: e^- -phonon scattering

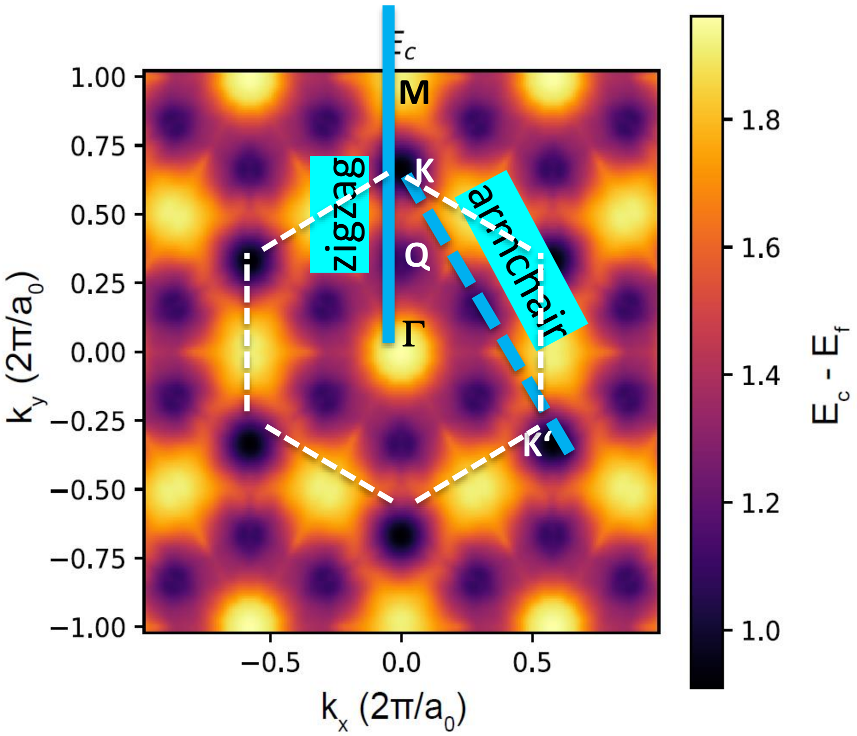


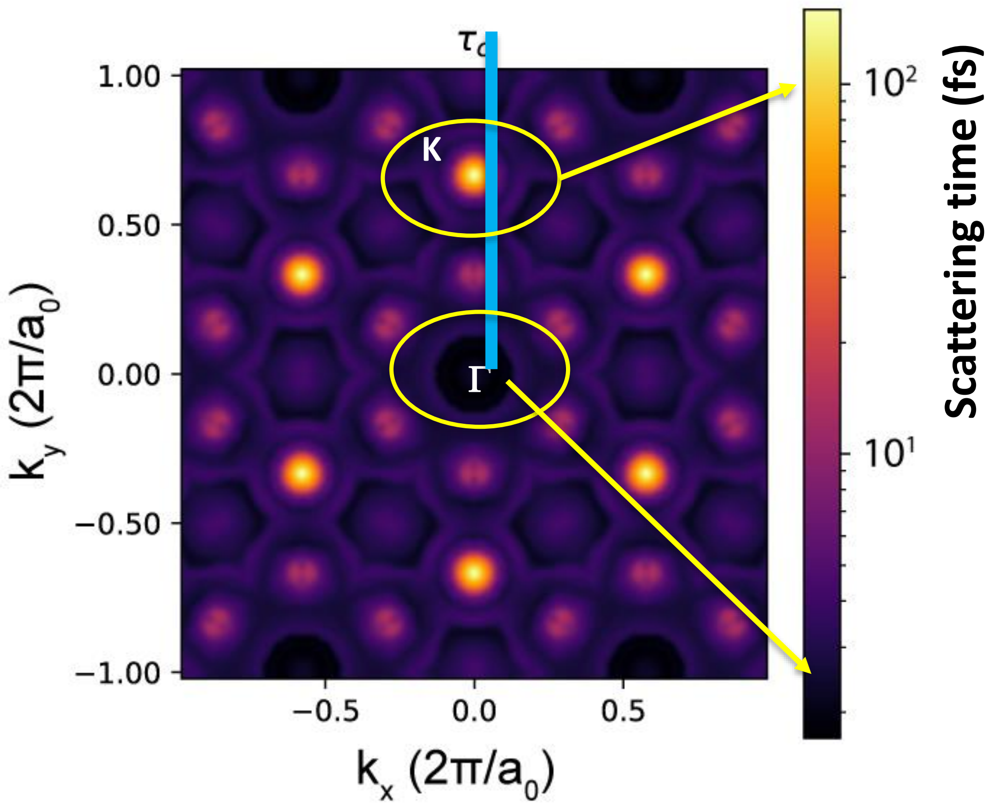


A) Electronic band structure and b) Phonon dispersion of WS_2

Origin of the dephasing: e^- -phonon scattering







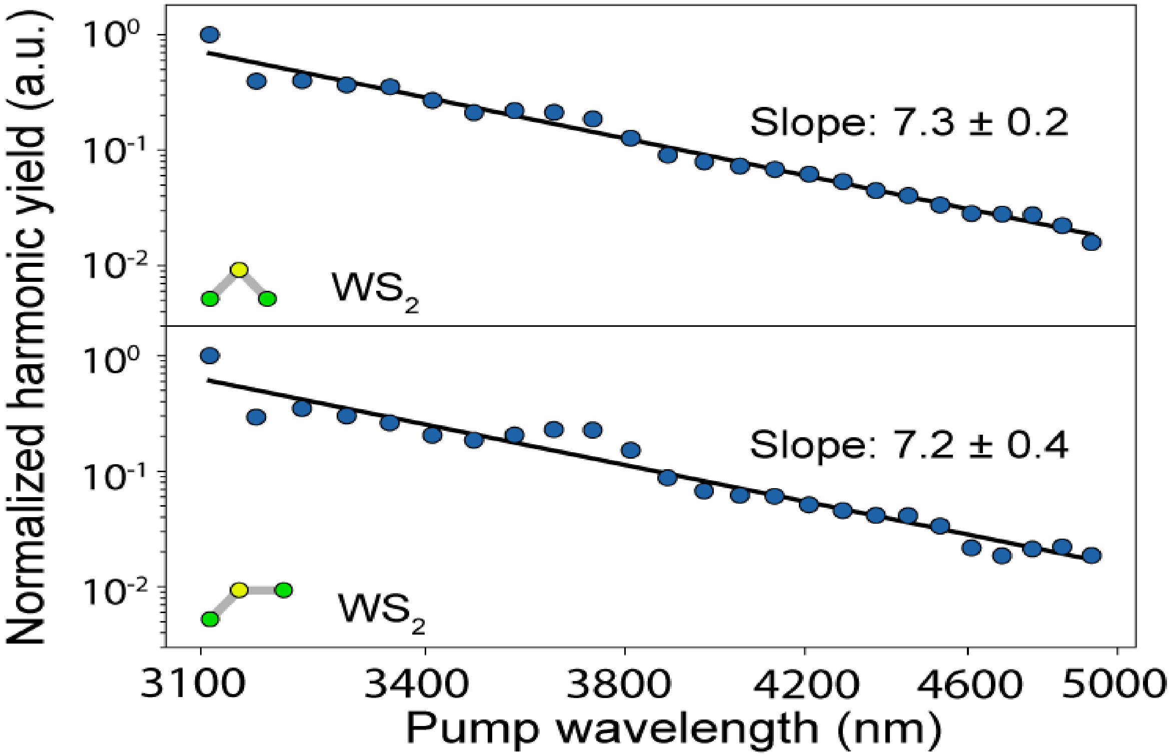
A) Electronic energy landscape (CB) and b) Calculated e^--phonon scattering time (LOG-scale!) in WS_2

- **e-phonon scattering time is highly dispersive → T_2(k)**
- **e-ph scattering time drops from ~200 fs in the K-valley down to ~2 fs in a vicinity of Γ-point!**

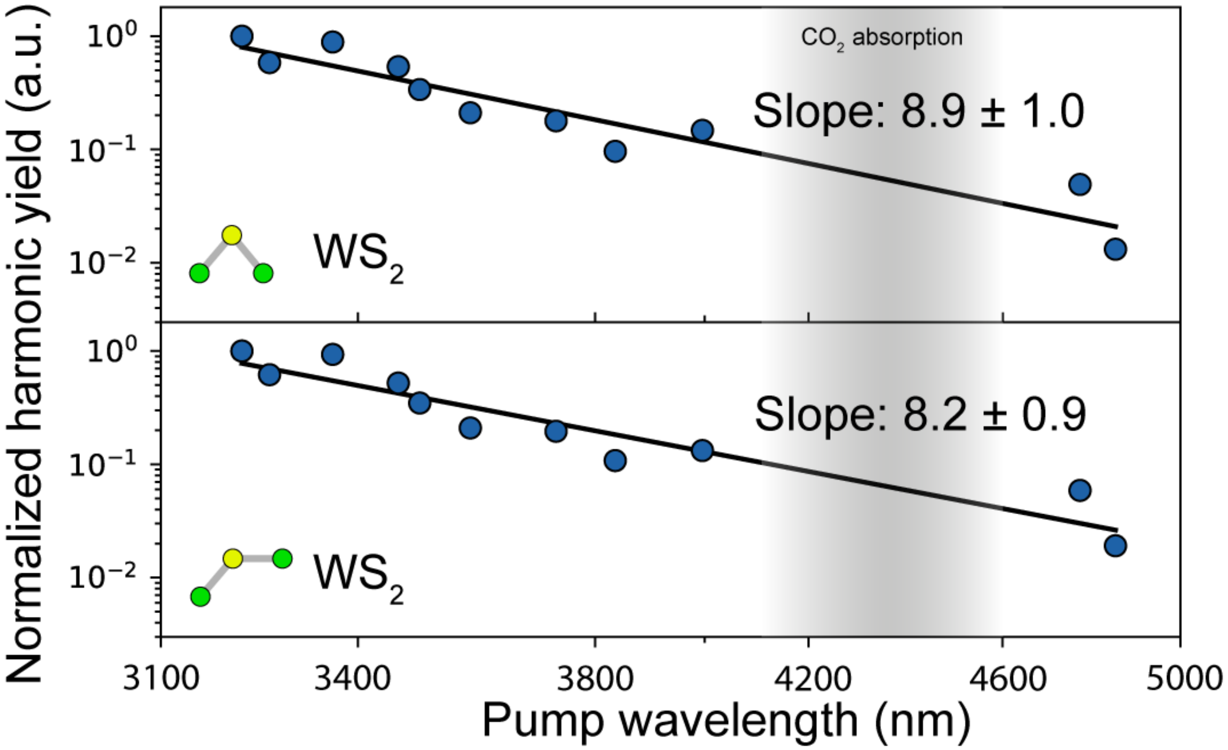
Numerical simulations

SBE simulations

rt-TDDFT simulations



Experiment



SBE simulations with ab-initio calculated $T_2(k)$-match the experimental results!

V. Korolev et al., „**Unveiling the Role of Electron-Phonon Scattering in Dephasing High-Order Harmonics in Solids", submitted; arXiv:2401.12929**

STRONG FIELD MEETS QUANTUM OPTICS

Quantum description of (semiconductor) HHG

NOA
NONLINEAR OPTICS
DOWN TO ATOMIC SCALES

Collaboration partners:

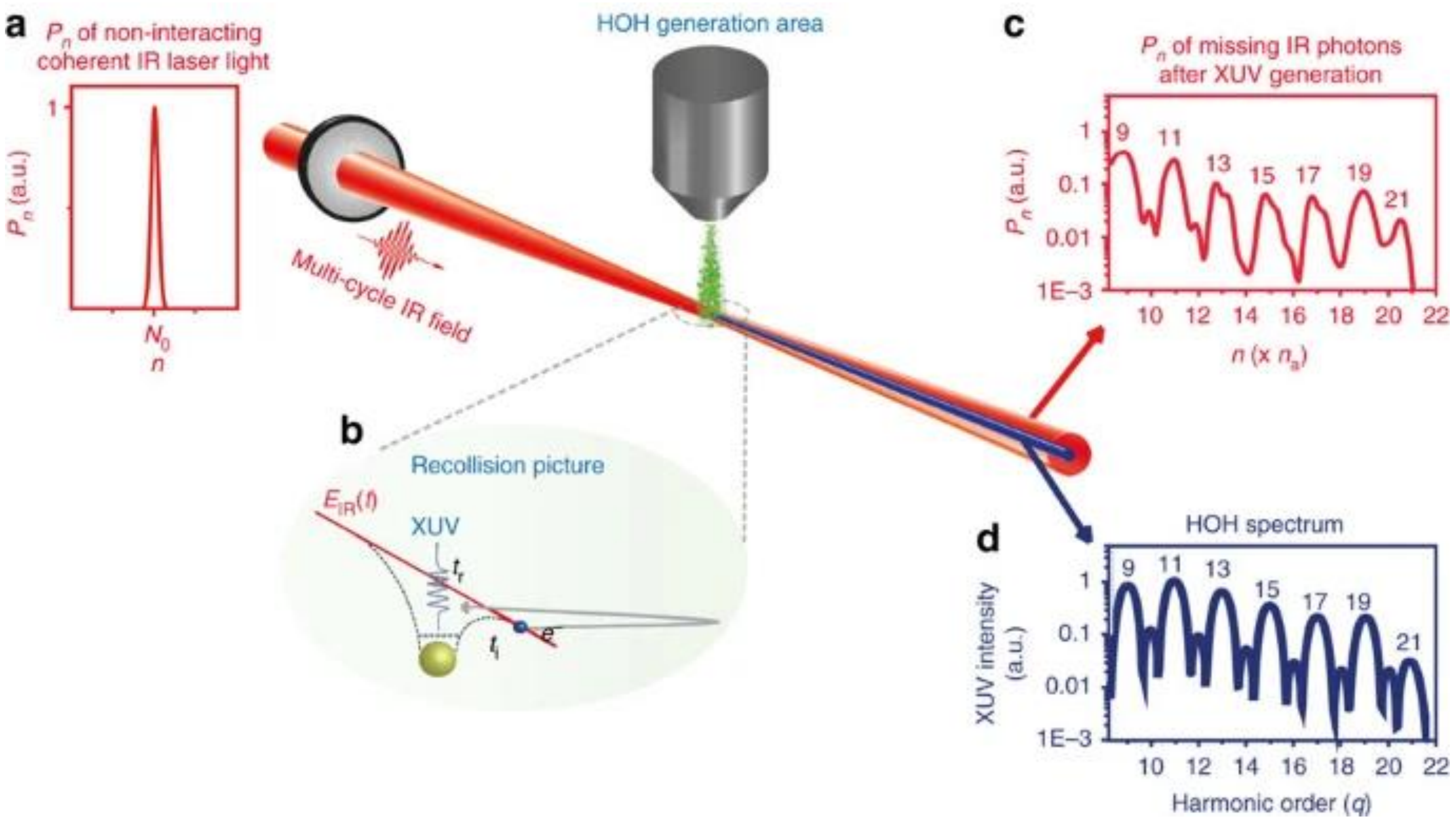
Prof. Jens Biegert (ICFO)

Prof. Ulf Peschel

Prof. Misha Ivanov (MBI Berlin)

Prof. Hamed Merdji (Paris)

Quantum Signatures in HHG – the pioneering experiment



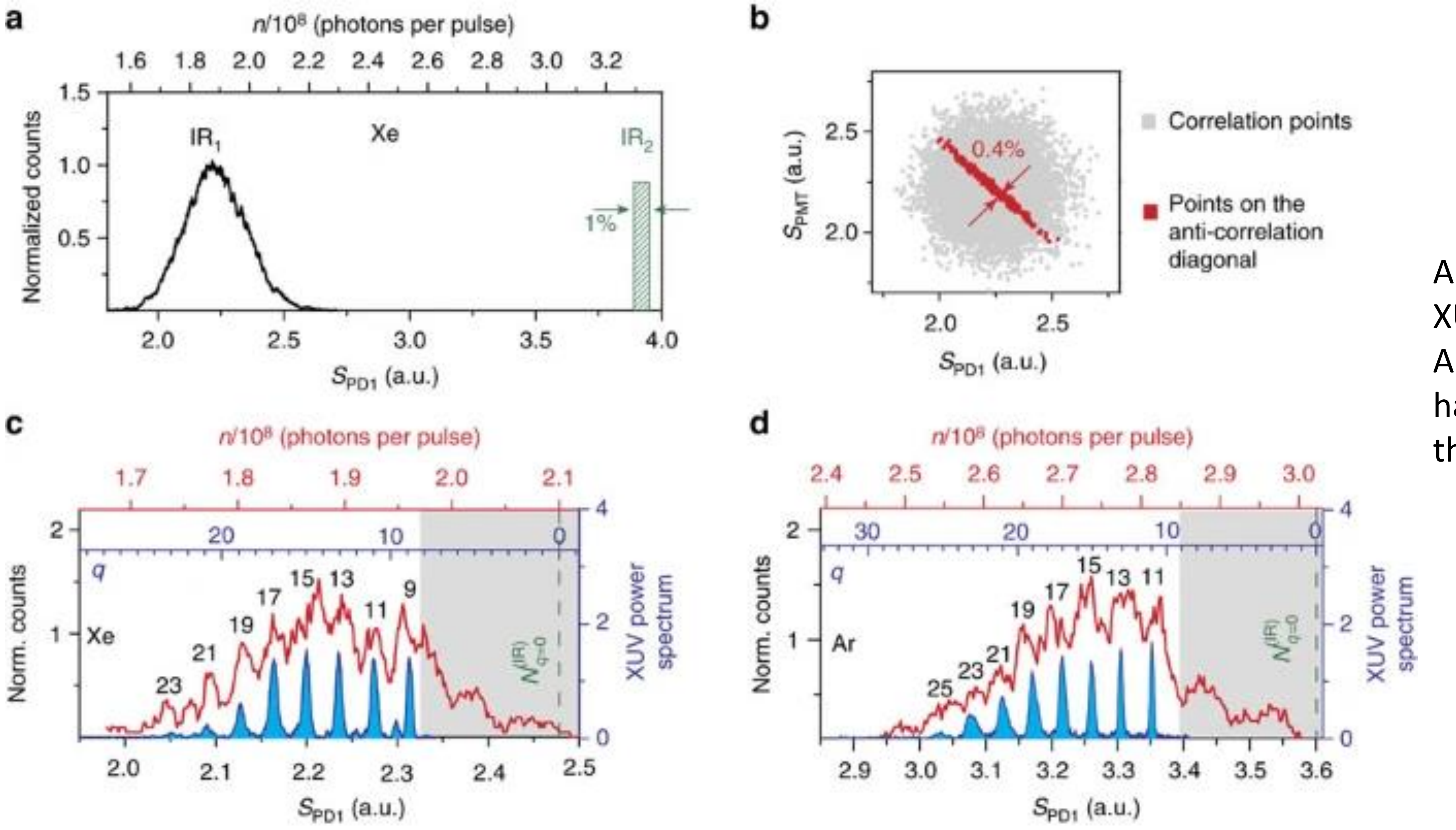
IR: 800 nm pulse, 0.6mJ energy/pulse; (~8x10^{13}W/cm^2), 10^{15} photons/pulse

Xe gas jet – generating HH

Afterwards: IR beam attenuated by ~10^6

N. Tsatrafyllis, I.K. Kominis, I. A. Gonoskov, P. Tzallas, Nat. Comm. 8, 15170 (2017)

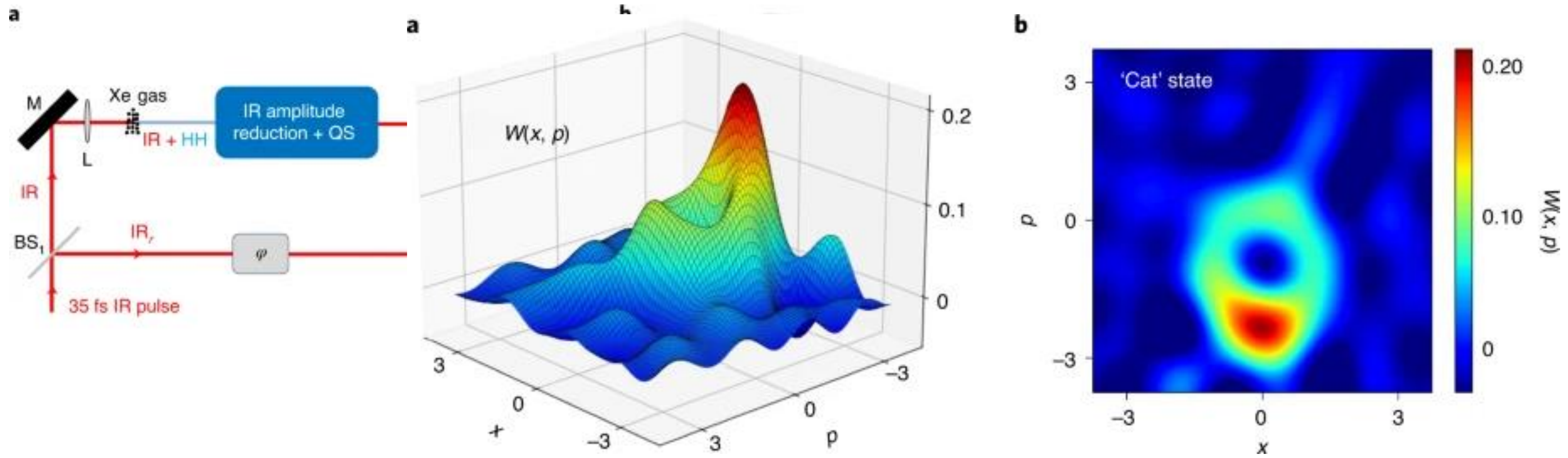
Quantum Signatures in HHG – the pioneering experiment



Apprxoimately 10^8 photons per XUV pulse,
About 10^7 photons per harmonics (for 5 harmonics in the plateau region)

N. Tsatrafyllis, I.K. Kominis, I. A. Gonoskov, P. Tzallas, Nat. Comm. 8, 15170 (2017)

Generation of optical „cat“ states in HHG

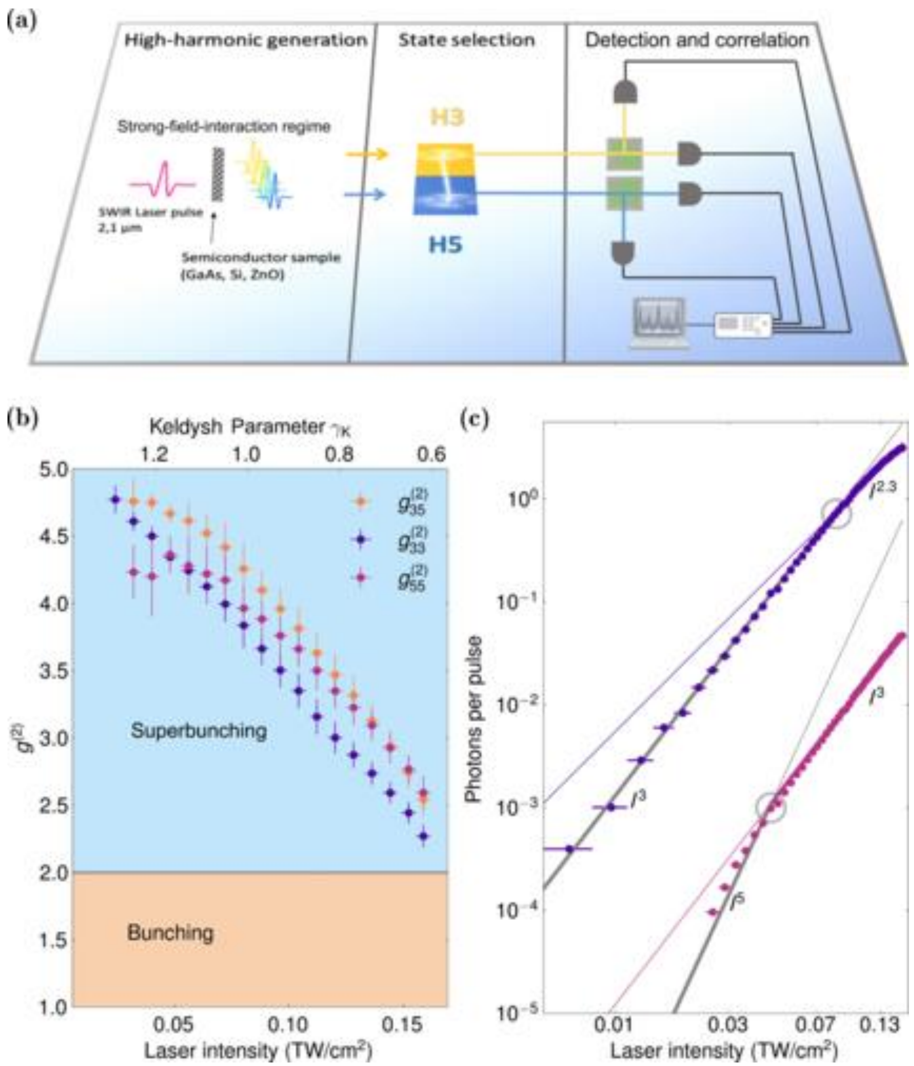


- Reconstruction of the transmitted fundamental radiation
- Nonclassicality (negative regions of the Wigner function)
- Obtained by conditioning

… so far: quantum properties of the transmitted (generating) fundamental (& to the HHs)

M. Lewenstein, M.F. Ciappina, E. Pisanty, J. Rivera-Dean, P. Stammer, Th. Lamprou, P. Tzallas, Nature Phys. 17, 1104 (2021)

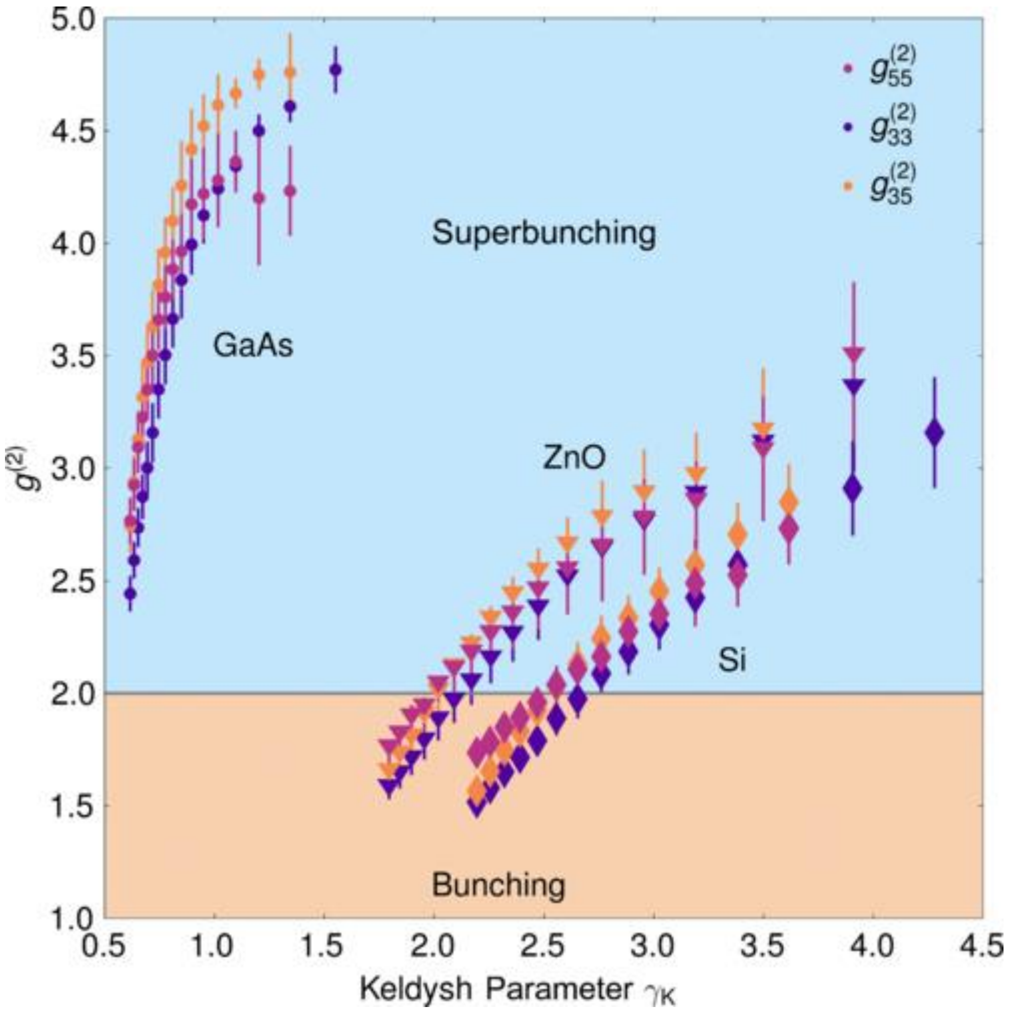
Quantum properties/entanglement of the harmonics



- HHG experiment in bulk semiconductors (GaAs Si, ZnO)
- Correlation $g^{(2)}$ measured ($g_{33}^{(2)}$, $g_{55}^{(2)}$ and $g_{35}^{(2)}$)
- $g^{(2)}$ changes as a function of laser intensity (Keldysh parameter)

D. Theidel, …U. Morgner, M. Kovacev, J. Biegert, H. Merdji, PRX Quantum 5, 040319 (2024)

Quantum properties/entanglement of the harmonics



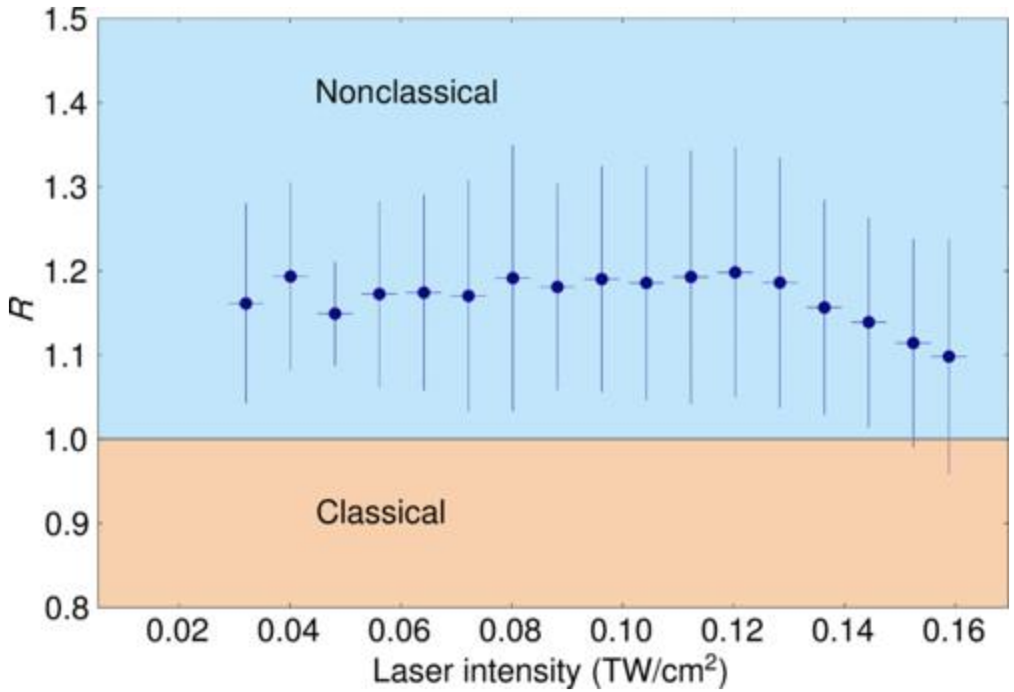
For a single-mode bosonic state:
$g^{(2)}$=1 – Poissonian photon-number statistics (coherent state).
$g^{(2)}$>1 – super-Poissonian statistics – bunched arrival of photons

$g^{(2)}$=2 – Bose-Einstein

$g^{(2)}$>2 Nonclassical effects such as superbunching,
$g^{(2)}<g^{(2)}(\tau)$ is connected to photon antibunching, as often obtained from single-photon sources.

D. Theidel, …U. Morgner, M. Kovacev, J. Biegert, H. Merdji, PRX Quantum 5, 040319 (2024)

Quantum properties/entanglement of the harmonics



Cauchy-Schwartz inequality

$$g_{ii}^{(2)} g_{jj}^{(2)} < \left| g_{ij}^{(2)} \right|^2$$

$$R := \frac{\left| g_{ij}^{(2)} \right|^2}{g_{ii}^{(2)} g_{jj}^{(2)}}$$

D. Theidel, …U. Morgner, M. Kovacev, J. Biegert, H. Merdji, PRX Quantum 5, 040319 (2024)

How to theoretically describe quantum optics in the strong-field regime?

Exact Factorization: general case

$$\mathrm{i}\,\dot{\Psi} = \hat{H}\,\Psi$$

$$\Psi(\mathbf{x},\mathbf{y},t) = \Phi(\mathbf{x},\mathbf{y},t)\cdot G(\mathbf{y},t)$$

$$\hat{H} = \left[\hat{H}_1(\mathbf{x}) + \hat{H}_2(\mathbf{y}) + \hat{W}_{int}(\mathbf{x},\mathbf{y},t)\right]$$

Abedi, A., Maitra, N. T., Gross, E. K. U., Phys. Rev. Lett. 105, 123002 (2010)

Exact Factorization: quantum optical description of laser-driven systems*

$$\Psi(\mathbf{x},\mathbf{y},t) = \Phi(\mathbf{x},\{\beta\mathbf{y}\},t)\cdot G(\mathbf{y},\beta,t) + O(\beta^s),$$

$$\Phi(\mathbf{x},\{\beta\mathbf{y}\},t) = F(\mathbf{x},\{\beta\mathbf{y}\},t)\cdot\exp\left[i\int^t \langle\hat{H}_1 + \hat{W}_{int}\rangle_F\,d\tau\right],$$

$$i\dot{F} = \left[\hat{H}_1 + \hat{W}_{int}(\mathbf{x},\{\beta\mathbf{y}\},t)\right]F,$$

$$i\dot{G} = \left[\hat{H}_2 + \langle\hat{H}_1 + \hat{W}_{int}\rangle_F\right]G.$$

Coordinate-scaling parameter $\beta \ll 1$

$$\beta \propto \sqrt{\frac{m}{M}},\quad \beta \propto \frac{1}{\sqrt{V_q}},\quad \ldots$$

*I. Gonoskov, S. Gräfe, "Light-matter quantum dynamics of complex laser-driven systems", *J. Chem. Phys.* *154*, 234106 (1-5) (**2021**).

Parametric Factorization: light-matter problem*

(Interaction representation, plane wave 1+1 case, zero initial phase, SF/coherent)

$$i\dot{\Psi} = \hat{H}\Psi\ , \quad \hat{H} = \frac{1}{2}\left(\hat{\vec{p}} - \hat{\vec{A}}\right)^2 + U(\vec{\mathbf{r}}, t)\ ,$$

$$\hat{\vec{A}} = -\mathbf{x_0}\{\beta q\}\ \cos(\kappa z - \omega t)$$

$$\Psi_0 = \Phi_0(\vec{\mathbf{r}}) \cdot G_c(q), \qquad N_0 \gg 1. \qquad A_0 = \beta\sqrt{2N_0}.$$

$$\beta = c\sqrt{2\pi/\omega V}$$

Coordinate-scaling parameter:

$$i\dot{F} = \left[\frac{1}{2}\left(\hat{\vec{p}} - \hat{\vec{A}}_{par}\right)^2 + U(\vec{r}, t)\right] F\ ,$$

$$i\dot{G} = \left\langle \frac{1}{2}\left(\hat{\vec{p}} - \hat{\vec{A}}\right)^2 + U(\vec{r}, t) \right\rangle_F G\ .$$

*I. Gonoskov, S. Gräfe, “Light-matter quantum dynamics of complex laser-driven systems”, *J. Chem. Phys*. *154*, 234106 (1-5) (**2021**).

How to theoretically describe quantum optics in the strong-field regime?

Evolution of the light quantum states under the back-action of the intraband current (interaction picture):

$$\frac{\partial}{\partial t}|G\rangle = n_e\, E_c\left(-\sum_j \frac{e}{c}\hat{\vec{A}}_j(t)\right)|G\rangle$$

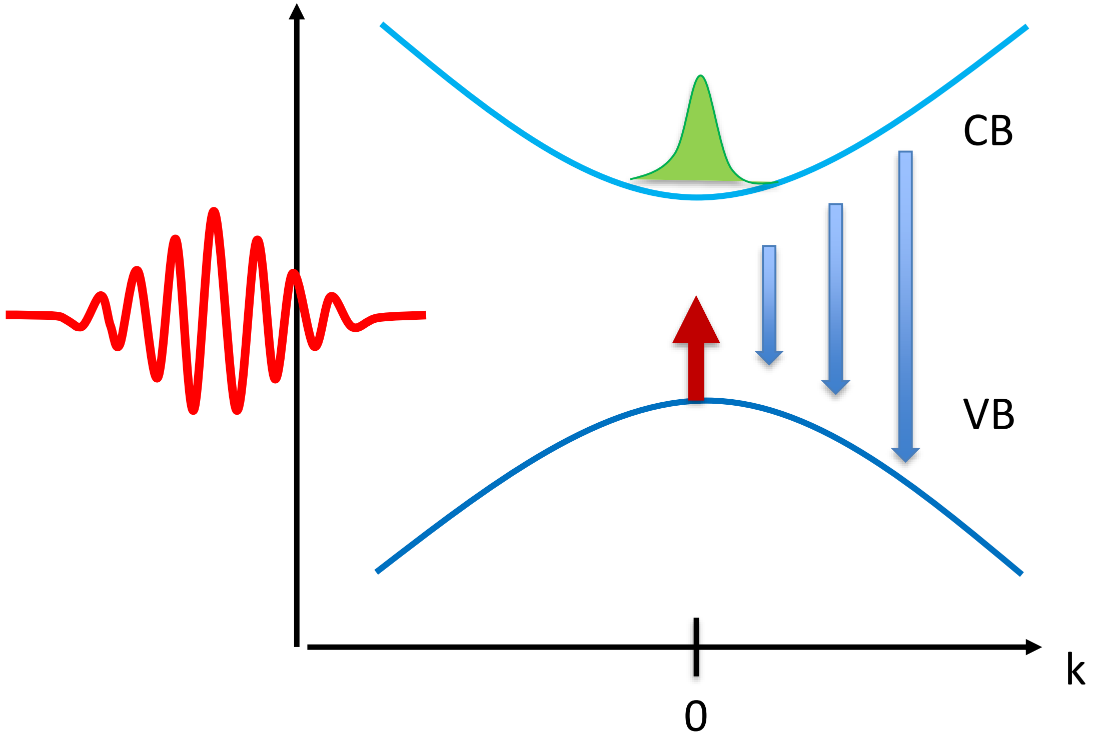
Vector potential operator:

$$\hat{A}(t) = \hat{A}_L(t) + \sum_{j\geq 2}\hat{A}_j(t)$$
$$= \sqrt{\frac{2\pi c^2}{\omega_L V}}\cos(\omega_L t)\,\hat{Q}_L + \sum_j \sqrt{\frac{\pi c^2}{\omega_j V}}\left[\hat{a}_j e^{-\omega_j t} + \hat{a}_j^\dagger e^{\omega_j t}\right].$$

$$i\frac{\partial}{\partial t}|G\rangle = \left[n_e\, E_c\left(\frac{e}{c}\hat{A}_L(t)\right) + n_e\sum_j \frac{e}{c}\hat{A}_j(t)\cdot\frac{\partial E_c}{\partial K}\Big|_{K=\frac{e}{c}\hat{A}_L}\right]|G\rangle$$

Non-classical light generation in semiconductor HHG

HHG in a semiconductor; intraband current contribution (below-bandgap harmonics)



Pauli Fierz Hamiltonian

$$\hat{H} = n_e \boxed{E_c} \left(\hat{\vec{p}} - \sum_j \frac{e}{c} \hat{\vec{\mathscr{A}}}_j \right) + \sum_j \omega_j \hat{N}_j,$$

Conduction band dispersion $E_C(k)$

Quantized electromagnetic field modes j

Approximation: quantum evolution of light field*, treating intraband-current back-reaction as a finite-order perturbation

*I. Gonoskov, S. Gräfe, "Light-matter quantum dynamics of complex laser-driven systems", *J. Chem. Phys. 154*, 234106 (1-5) (**2021**).

I. Gonoskov, R. Sondenheimer, …S. Gräfe, „Nonclassical light generation and control from laser-driven semiconductor intraband excitations“, Phys. Rev. B *109*, 125110 (**2024**)

Non-classical light generation in semiconductor HHG

Separation: radiation field and field-dressed semiconductor

Initial state of light: product of coherent state (fundamental of the laser) and vacuum states (harmonic modes upto cutoff)

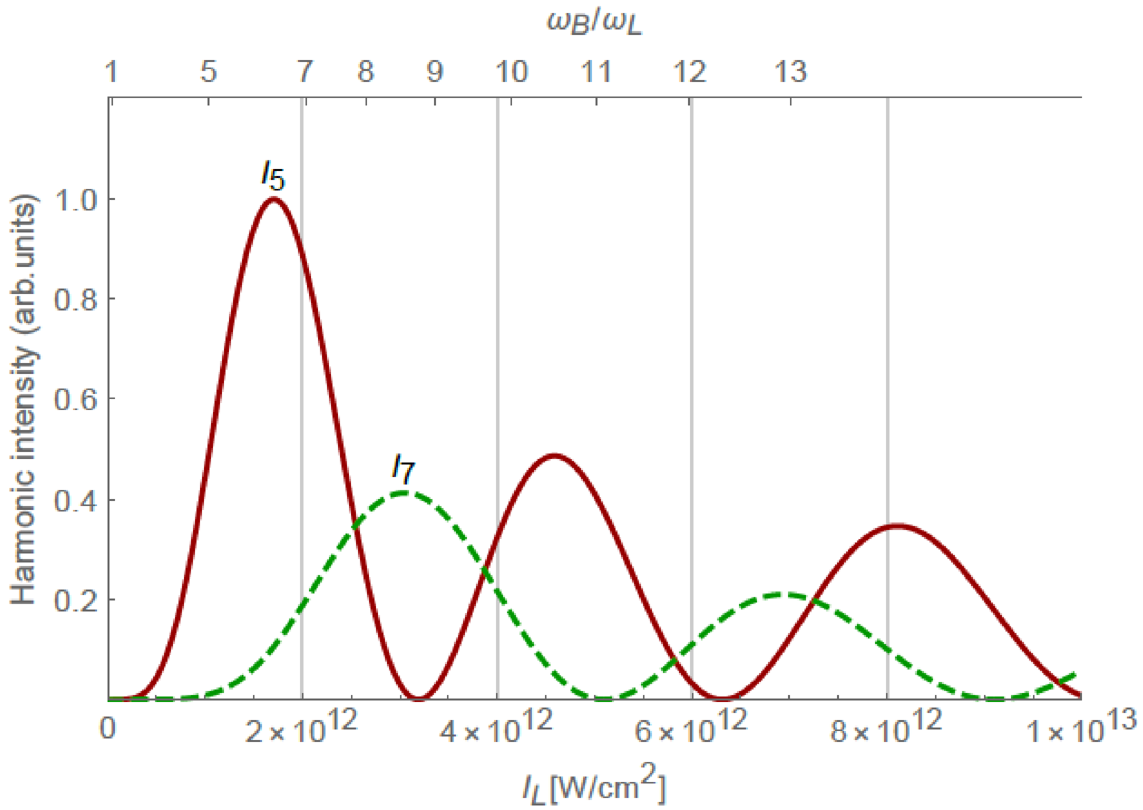
$$|G >= |a_{Laser} > \otimes |0_j >$$

$$\mathrm{i}\frac{\partial}{\partial t}|G\rangle = \left[n_\mathrm{e} E_\mathrm{c}\left(\frac{e}{c}\hat{A}_\mathrm{L}(t)\right) + n_\mathrm{e}\sum_j \frac{e}{c}\hat{A}_j(t)\cdot\frac{\partial E_\mathrm{c}}{\partial K}\bigg|_{K=\frac{e}{c}\hat{A}_\mathrm{L}}\right]|G\rangle\,.$$

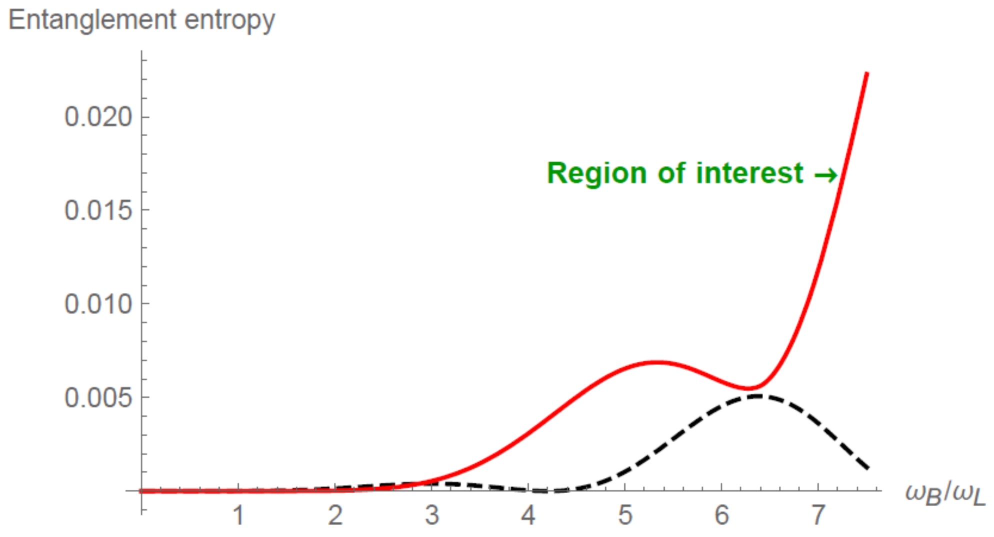
Equation is linear with respect to non-local operators (momentum quadrature operators) – analytical solutions

I. Gonoskov, R. Sondenheimer, …S. Gräfe, „Nonclassical light generation and control from laser-driven semiconductor intraband excitations“, Phys. Rev. B *109*, 125110 (**2024**)

Nonclassical properties of semiconductor HHG



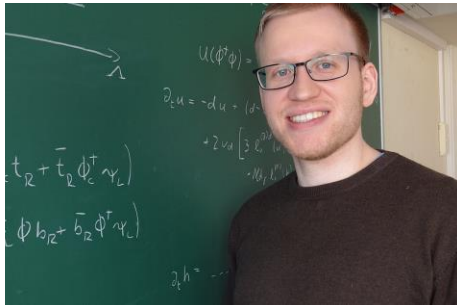
$$\mathrm{G}\sim\, G_0(\vec{Q})\, \mathrm{e}^{\delta_3 Q_L Q_3}\, \mathrm{e}^{\delta_5 Q_L Q_5}\, \ldots,$$



Many open questions

- Why are there actually quantum signatures in such intense driving fields? Should be all classical? Quantum character due to measurement (conditioning) or intrinsic?
- How to experimentally access ‘quantumness’? Interferometric measures?
- How to properly characterize bright, entangled squeezed quantum light (von Neumann entropy not an ideal measure)?
- Why does ‘quantumness’ surveil? So much decoherence everywhere… (and how to properly include decoherence…?)

- Numerical model (incl. dephasing time)
- Are there some ‘sweet spots’ for harnessing quantum light?
- What about squeezed light?



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NOA NONLINEAR OPTICS DOWN TO ATOMIC SCALES

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Thank you for your attention!

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