ELI-ALPS Research Institute
TOWARDS THE SHARP END OF ATTOSCIENCE
eli attosecond

High Harmonic Generation: A Computer Experiment

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September 15, 2022

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photon and neutron open science cloud

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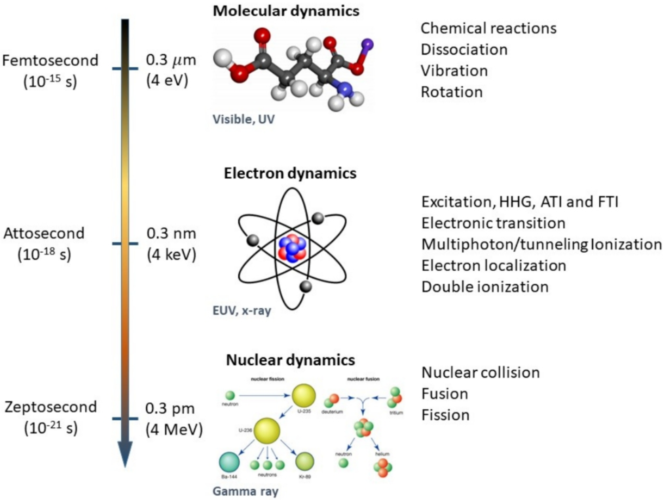
Ultrafast Science



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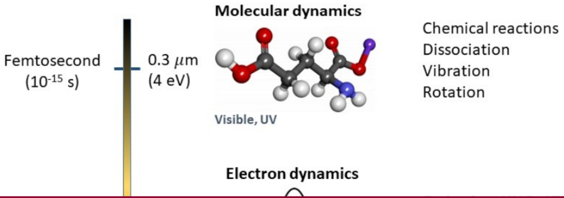
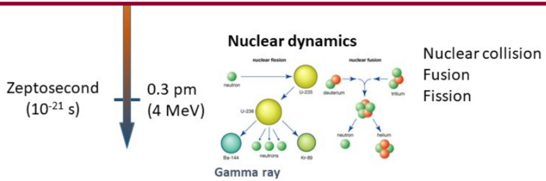


Ultrashort pulses

- What is an ultrashort pulse?



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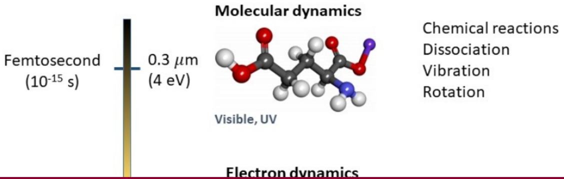
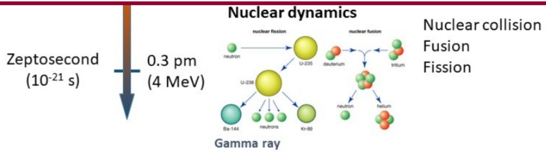


Ultrashort pulses

- What is an ultrashort pulse?
- How to generate it?



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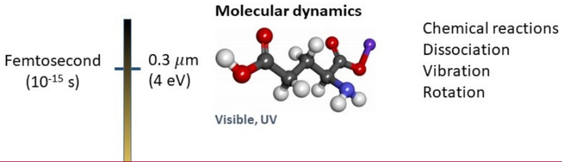
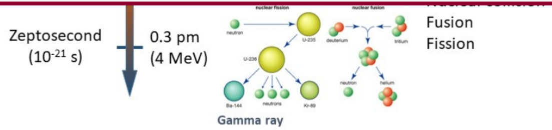


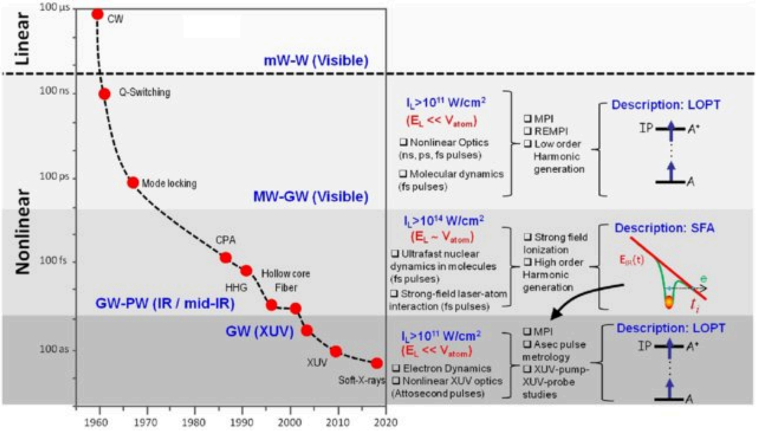
Ultrashort pulses

- What is an ultrashort pulse?
- How to generate it?
- How to measure it?



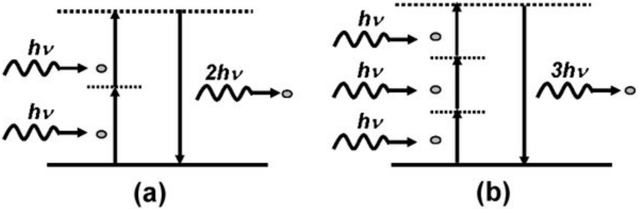
https://corels.ibs.re.kr/html/corels_en/research/research_0305.html

Pulse duration

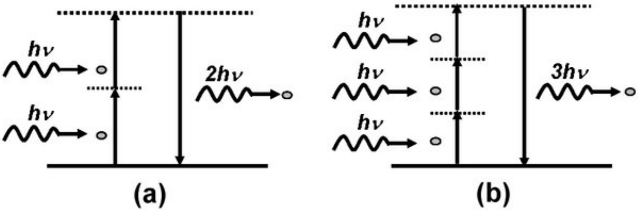


I. Orfanos et al., APL Photonics 4, 080901 (2019)

Harmonics Generation



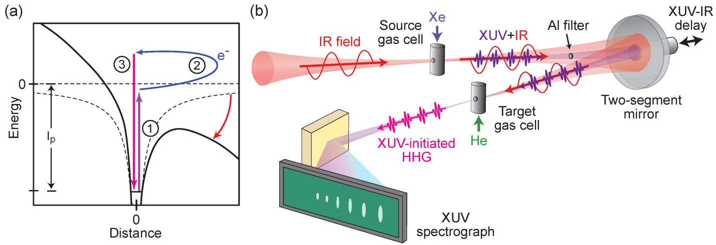
M. R. Tsai et al., J. of Biomedical Optics, 15(2), 026002 (2010).



Classical harmonic generation with few photons
Low efficiency
Perturbative non-linear optics
Low order harmonics

M. R. Tsai et al., J. of Biomedical Optics, 15(2), 026002 (2010).

High Harmonics Generation (HHG)

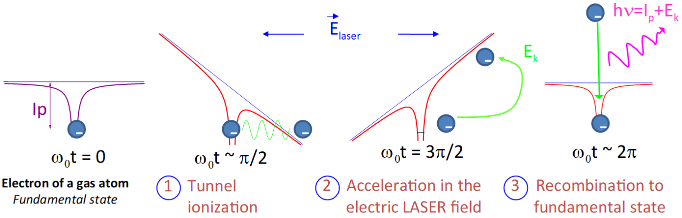


Three step model:

1. Tunnel ionization — Involves several photons
2. Acceleration — Non-perturbative process
3. Recombination — High-order harmonics

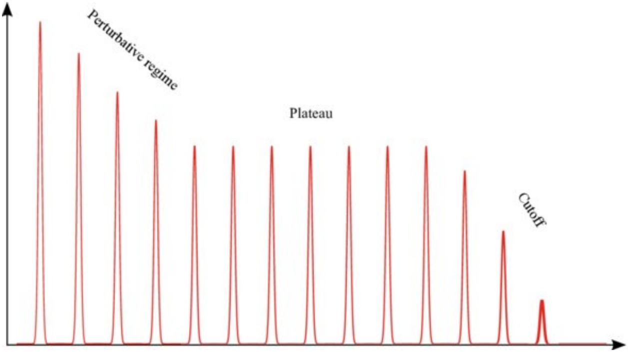
P.B. Corkum PRL 71, 1994 (1993)
K. Kulander et al. SILAP (1993)
M. Krüger et al., Appl. Sci. 2019, 9(3), 378.

Semi-Classical model



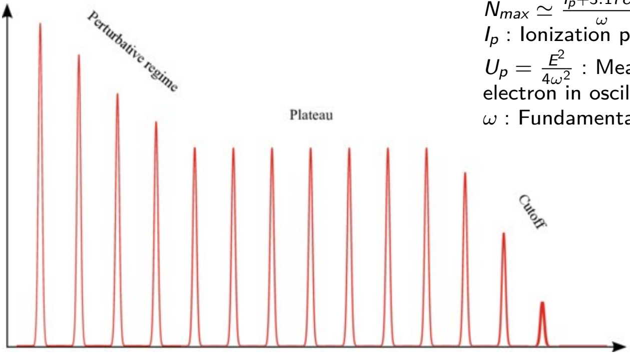
Adapted from presentation by Benoît MAHIEU
https://slideplayer.com/slide/4173468/

Cutoff Law



Hort, Ondrej. High harmonic generation with high energy femtosecond pulses . Diss. Bordeaux, 2014.

Cutoff Law



$N_{max} \simeq \frac{I_p + 3.17 U_p}{\omega}$

I_p : Ionization potential

$U_p = \frac{E^2}{4\omega^2}$: Mean KE acquired by electron in oscillating laser field

ω : Fundamental laser frequency

Hort, Ondrej. High harmonic generation with high energy femtosecond pulses . Diss. Bordeaux, 2014.

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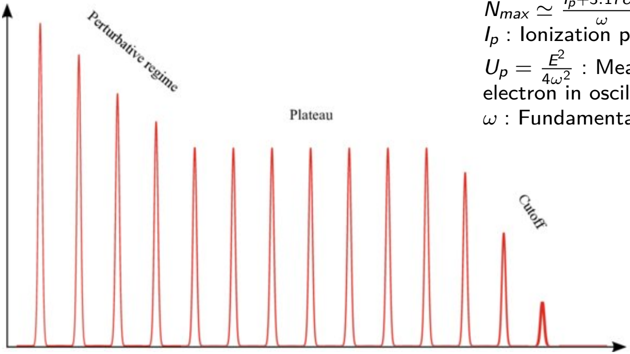

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I_p : Ionization potential

$U_p = \frac{E^2}{4\omega^2}$: Mean KE acquired by electron in oscillating laser field

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Keldysh parameter (γ): Distinguish perturbative interaction regime from non-perturbative regime $\left(\gamma = \sqrt{\frac{I_p}{2U_p}}\right)$.

$\gamma \ll 1$: Perturbative tunnelling OR over-the-barrier ionization regime

$\gamma \gg 1$: Non-perturbative multi-photon ionization

Hort, Ondrej. High harmonic generation with high energy femtosecond pulses . Diss. Bordeaux, 2014.

Trajectory and KE

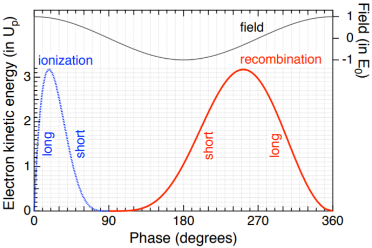


Fig. 2. Electron kinetic energy just before recombination normalized to the ponderomotive energy $E_{kin}(\theta_r)/U_p$ as a function of phase of ionization θ_i and recombination θ_r. The laser field normalized to the field amplitude $E(t)/E_0$ is also plotted in thin solid line (right axis).

Maximum value for kinetic energy of electron ($3.17U_p$) achieved when $\theta_i = 17°$ and $\theta_r = 255°$

http://ishiken.free.fr/Publication/intech.pdf

Theory: Lewenstein Integral

In 1994 Lewenstein et al. solved the time dependent Schrödinger equation (TDSE) in a single-electron approximation under influence of linearly polarized laser field Ecos(t).
In length gauge the TDSE takes the form:

$$i\left|\Psi(x,t)\right\rangle = \left[-\frac{1}{2}\nabla^2 + V(x) - Ecos(t)x\right]\left|\Psi(x,t)\right\rangle$$

$V(r)$: Atomic potential
Non-linear dipole response D(t) using the dipole operator $\hat{D}$:

$$D(t) = \left\langle\Psi(t)\right|\hat{D}\left|\Psi(t)\right\rangle$$

$$D(t) \approx -j\int_{t_0}^{t} dt' \int dpe^{-j\int_{t'}^{t} d\tau[\frac{[p+A(\tau)]^2}{2}]+I_p}$$

Lewenstein, Maciej, et al. Physical Review A 49.3 (1994): 2117.

Theory: Lewenstein Integral

For a single atom case, $\hat{D} = -\hat{r}$ and $t_0 \to -\infty$
Lewenstein Integral:

$$D(t) \approx j \int_{-\infty}^{t} dt' \int dp \underbrace{e^{-jI_p t} d^*[p + A(t)]}_{Recombination} \times \underbrace{e^{-jS_{GV}(p,t,t')}}_{Propagation} \times \underbrace{e^{jI_p t'} F(t') d[p+A(t')]}_{Ionization} + c.c.$$

Dipole matrix elements: $d[p + A(t')] = \langle p + A(t') | \hat{r} | g \rangle$ and $d^*[p + A(t')]$
Vector potentials: $A(t')$ and $A(t)$

Lewenstein, Maciej, et al. Physical Review A 49.3 (1994): 2117.

Theory: TDDFT

In TDDFT method, using Runge-Gross theorem a one-to-one correspondance between the time dependent potentials and densities is established. Hence the TDKS equation is given by:

$$i\frac{\partial}{\partial\, t}\phi_i(r,t) = [-\frac{\nabla^2}{2} + V_{ks}[n](r,t)]\phi_i(r,t)$$

$V_{ks}[n](r,t) = V_{ext}(r,t) + V_H[n](r,t) + V_{XC}[n](r,t)$:
KS potential given by one body external potential, non-interacting Hartree potential and XC potential.

$V_H[n](r,t) = \int dr' \frac{n(r',t)}{|r-r'|}$: Hartree potential

$\phi_i(r,t)$: KS orbital

$n(r,t) = \sum_i^N |\phi_i(r,t)|^2$: Time dependent densiyt of N-interacting systems

Dobson, John F., M. J. Bünner, and E. K. U. Gross. Physical review letters 79.10 (1997): 1905.
Chu, Xi, and Patrick J. Memoli. Chemical Physics 391.1 (2011): 83-87.

Theory: TDDFT

Time dependent dipole moment:

$$d(t) = \int dr\ rn(r,t) = \sum_j \int dr\ r|\phi_j(r,t)|^2$$

Acceleration dipole moment:

$$a(t) = \frac{d^2}{dt^2} d(t) = -\int dr\ n(r,t)\nabla\ V_{ks}(r,t)$$

Fourier transform of the induced dipole moment modulus squared: harmonic generation dipole power spectrum.

$$P(\omega) = \frac{1}{t_f^2}\left|\int_0^{t_f} dt\ e^{-i\omega\ t} d(t)\right|^2$$

Fourier transform of the acceleration dipole moment: harmonic spectrum in the direction of polarization

$$H(\omega) = \frac{1}{t_f^2}\left|\int_0^{t_f} dt\ e^{-i\omega\ t} a(t)\right|^2$$

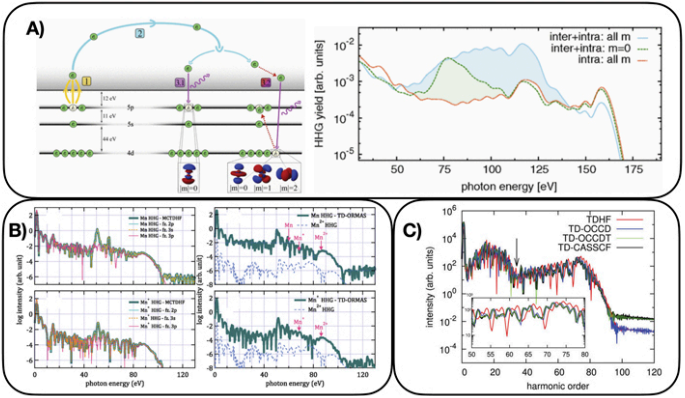
Dobson, John F., M. J. Bünner, and E. K. U. Gross. Physical review letters 79.10 (1997): 1905.
Chu, Xi, and Patrick J. Memoli. Chemical Physics 391.1 (2011): 83-87.

Approximations

- **Single active Electron Approximation**: Only one electron actively participates and electron–electron correlations are neglected.
- **Electric Dipole Approximation**: The driving field is spatially homogeneous over the spatial length scale relevant for the interacting system ($\hat{V}_L(\hat{r}, t) \rightarrow \hat{V}_L(t)$).
- **No Resonances**: $I_p \gg \omega_0$
- **Strong Field Approximation**: The electron propagating in the continuum interacts only with the laser field and the parent ion's potential is neglected.
- **Quasi-Static Approximation**: Tunnelling ionization dominant.
- **Weak Ionization Limit**: Driving laser intensity is less that the saturation intensity of the driven system.

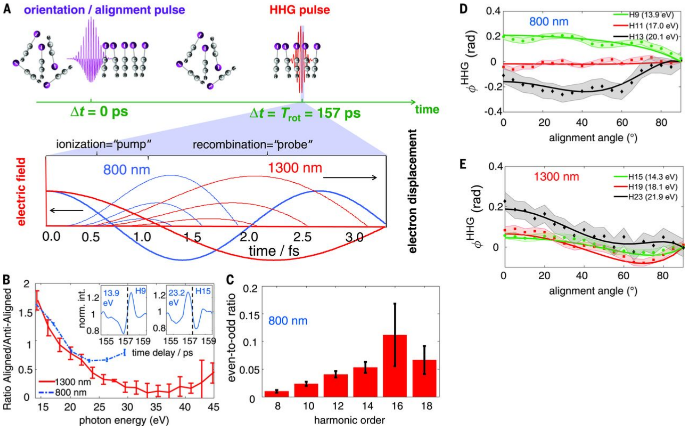
Nayak, Arjun, et al. Physics Reports 833 (2019): 1-52.

Examples: Different methods



E. Coccia and E. Luppi 2022 J. Phys.: Condens. Matter 34 073001
Sato T, Pathak H, Orimo Y and Ishikawa K L 2018 J. Chem. Phys. 148 051101
Pabst S and Santra R 2013 Phys. Rev. Lett. 111 233005
Wahyutama I S, Sato T and Ishikawa K L 2019 Phys. Rev. A 99 063420

Examples: Charge migration



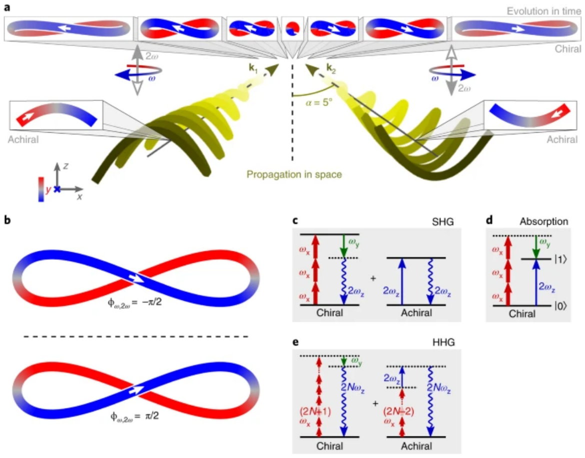
Kraus, Peter M., et al., Science 350.6262 (2015): 790-795.

Examples: Charge migration

- Generate HHS for the molecule of interest at **different orientation**.
- Take **ratios of the emission intensity** between molecules aligned perpendicular or parallel to the polarization of the probe pulse.
- Experimental **intensity ratios and phase differences**.
- The initial and final populations and phases were retrieved in a global nonlinear least-squares optimization using a **Levenberg-Marquardt algorithm** with multiple starting values.
- IMPORTANT: *All relevant electronic states*, the continuum structure through the use of scattering-wave matrix elements, nuclear motion through autocorrelation functions derived from photoelectron spectra, and the molecular axis distribution.
- The *photo-recombination dipole matrix* elements and the angular variation of the ionization rates can be calculated **theoretically**.
- These quantities are **experimentally** accessible using narrow-band extreme ultraviolet sources and/or charged-particle detection.
- The mapping from photon energy to transit time (t=t–t) can be performed using quantum electron trajectories obtained by the **saddle-point method**.

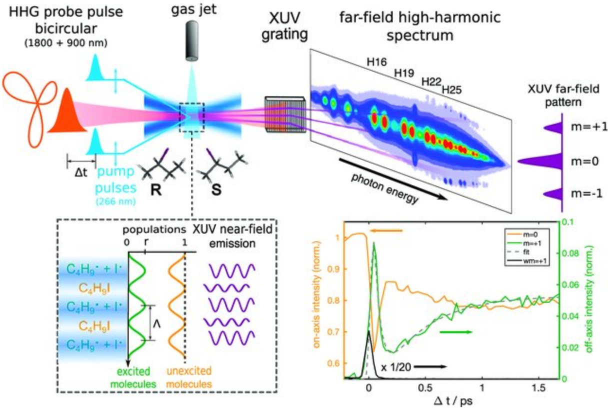
Kraus, Peter M., et al., Science 350.6262 (2015): 790-795.

Examples: Chiral molecules



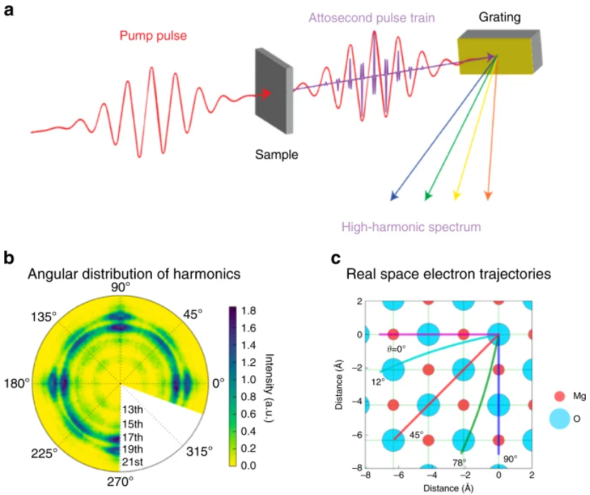
Ayuso, David, et al., Nature Photonics 13.12 (2019): 866-871.

Examples: Chiral molecules



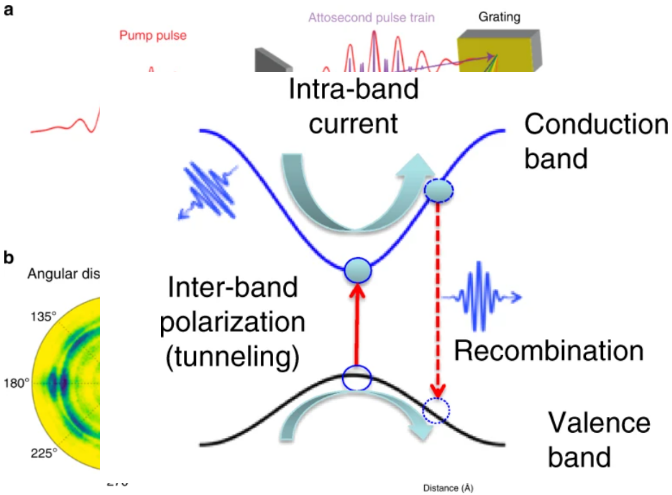
Baykusheva, Denitsa, et al. Proceedings of the National Academy of Sciences 116.48 (2019): 23923-23929.

Examples: Other



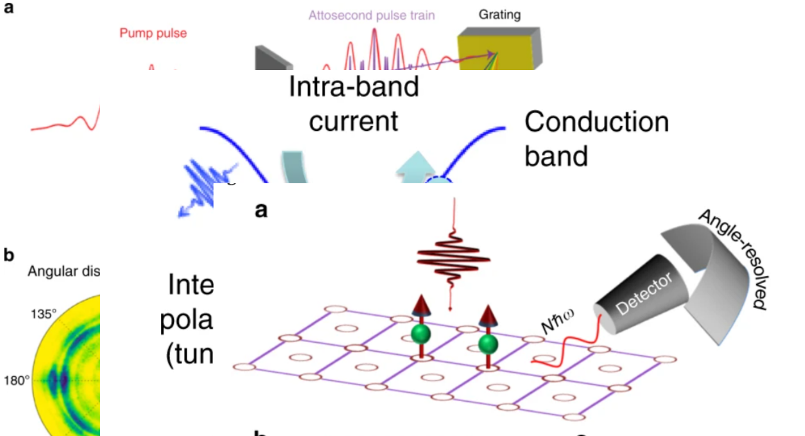
Y. You et. al., Nat. Phys. 13, 345–349 (2017).

Examples: Other



Y. You et. al., Nat. Phys. 13, 345–349 (2017).
S. Ghimire et. al., Nat. Phys. 15, 10–16 (2019).

Examples: Other



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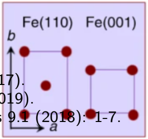

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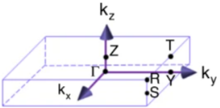
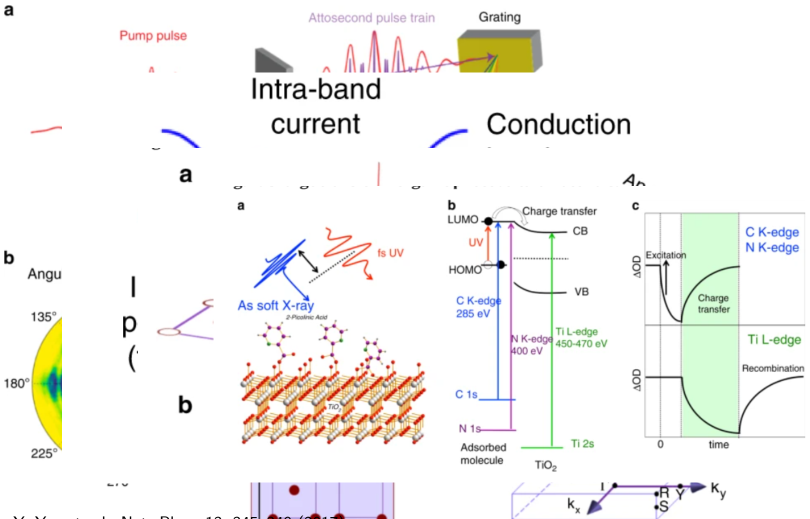


Y. You et. al., Nat. Phys. 13, 345–349 (2017).
S. Ghimire et. al., Nat. Phys. 15, 10–16 (2019).
Zhang, G. P., et al. Nature communications 9.1 (2018): 1-7.

Examples: Other



Y. You et. al., Nat. Phys. 13, 345–349 (2017).
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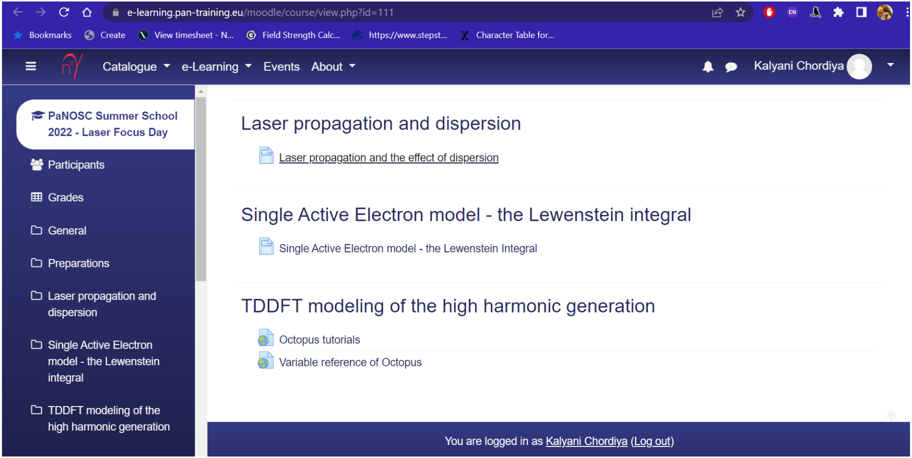
Zhang, G. P., et al. Nature communications 9.1 (2018): 1-7.
Li, Jie, et al. Nature Communications 11.1 (2020): 1-13.

Tutorial: Laser Day



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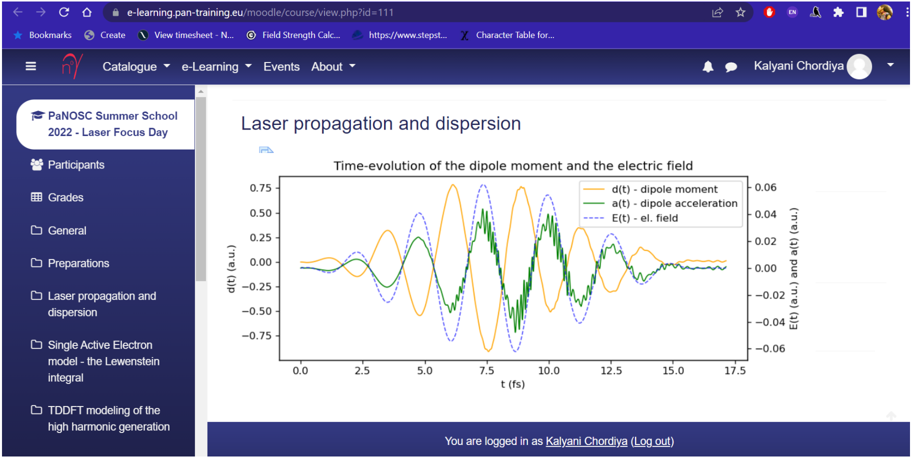

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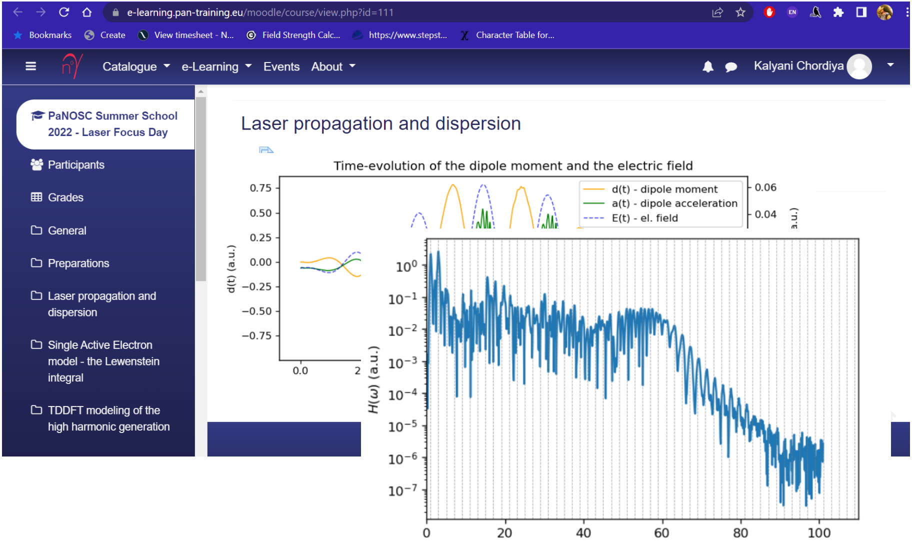

Summary and Take home

- Need for HHG in ultrafast science
- Steps in HHG process
- Models used to study HHG
- Theoretical methods for simulating HHG
- Application of HHG in studying electronic and structural dynamics
- Overview of the tutorial for this evening.

Acknowledgement

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