SENDISO

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Coherent control of light-driven electrons in solids

A milestone for multiqubit computing?

Thibault J.-Y. Derrien[1,2]

[1] Laboratory of Quantum Computing, IT4Innovation, VŠB Technical University of Ostrava, Czech Republic,
[2] Department of Scientific Laser Applications, HiLASE Centre, FZU Institute of Physics (AS CR), Dolní Břežany, Czech Republic

Quantum computing seminar EuroCC,
September 26th, 2024.

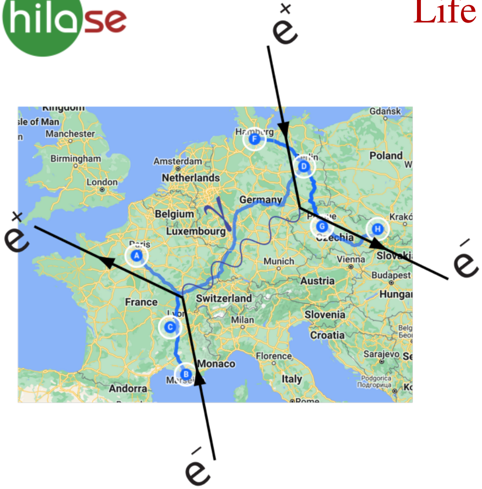
hilase FZU

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 739573 (HiLASE CoE)

EUROPEAN UNION
European Structural and Investment Funds
OP Research,
Development and Education

MŠMT UKRI Science and Technology Facilities Council

Life scattering diagram



Thibault J.-Y. Derrien, French, 39 years old.

2008. Master degree Plasma Physics (U. Paris-Saclay/Ecole Polytechnique, France).

2012. PhD degree, Aix-Marseille University, LP3 / CNRS, Marseille (France).

2012. Lab. Hubert Curien LabHC / CNRS, St-Etienne (France).

2013. BAM Fed. Inst. Mat. Res. Test., Berlin (Germany).

2015. Marie Curie Individual Fellow ”QuantumLaP” at HiLASE Prague (Czech Republic).

2017. Senior researcher @ FZU (Prague)

2018. Post-doc at **Max Planck Institute** (MPSD Hamburg, Germany)

2019. Marie Curie RISE ”ATLANTIC” networking program (Prague, Czech Republic)

2021. **Group lead** ”Ultrafast photonics” @ FZU Prague.
2024. Group lead ”Quantum Dynamics of Systems” @ IT4I Ostrava.

15 years research experience

Stayed (0.5 - 10) years in 9 EU research laboratories, multi-cultural environments, 2/3rd in experimental groups.

Bibliometrics

Thibault J.-Y. Derrien

FOLLOWING

HiLASE Centre, FZU Institute of Physics, Czech Academy of Sciences
Verified email at fzu.cz - Homepage

Laser nanostructures plasmonics density functional theory non-linear dynamics

TITLE	CITED BY	YEAR
High-speed manufacturing of highly regular femtosecond laser-induced periodic surface structures: physical origin of regularity I Gnilitskyi, TJY Derrien, Y Levy, NM Bulgakova, T Mocek, L Orazi Scientific reports 7 (1), 8485	328	2017
Fundamentals of ultrafast laser–material interaction MV Shugaev, C Wu, O Armbruster, A Naghilou, N Brouwer, DS Ivanov, ... MRS Bulletin 41 (12), 960-968	248	2016

Cited by VIEW ALL

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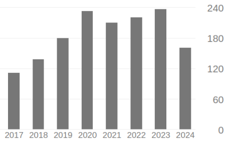
Citations	1735	1243
h-index	17	16
i10-index	24	21



Web Of Science

- H-index: 15.
- Publications: 39 (WOS). 2 patents (CZ, EU). 3 chapters in monographs.
- Number of citations: ∼1,275 (WOS).

Acknowledgments & funding

09-2024 **IT4Innovation**

2024 **Ministry of Education Youth & Sports** - CZ-MSMT-OPJAK ”**Sendiso**” for developing sensors for biology using laser technologies (”Sensors and Detectors for the Future Information Society”).

2017-12/2023 **European Regional Development Fund** and the **state budget of the Czech Republic** (project **BIATRI**: CZ.02.1.01/0.0/0.0/15_003/0000445, **project HiLASE CoE**: No. CZ.02.1.010.00.015_0060000674).

2019-2024 **EU-H2020-MSCA-RISE** European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 823897. Project ”ATLANTIC” (2019-2024).

2019-2024 **IT4Innovations** National Supercomputing Center – e-INFRA CZ (ID:90140) projects. National Grid Infrastructure **MetaCentrum** (CESNET LM2015042).

EUROPEAN UNION
European Structural and Investment Funds
Operationnal Programme Research,
Development and Education

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 739573 (HiLASE CoE)

<thibault.derrien@vsb.cz>

2019-2024: ATLANTIC network

ATLANTIC-RISE

Advanced theoretical network for modeling light matter interaction

News · Project summary · Participants · **Secondments** · Publications · Links

Secondments

Map of the secondments

FUNDING

This research is funded by the Marie Sklodowska-Curie Actions (MSCA) Research and Innovation Stack Exchange (RISE) of the European Union (EU) under Grant Agreement No. 823897.

Search ...

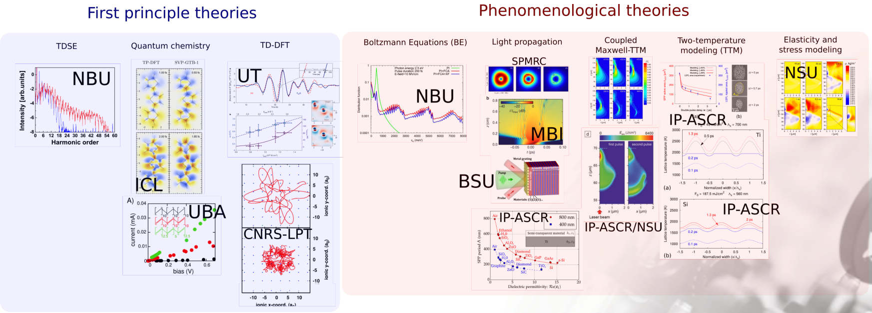
RECENT POSTS

- Secondment eligibility test
- CNRS-CELIA is leaving the Consortium for

https://www.QuantumLap.eu/?s=ATLANTIC

2019-2024: Marie Curie RISE network

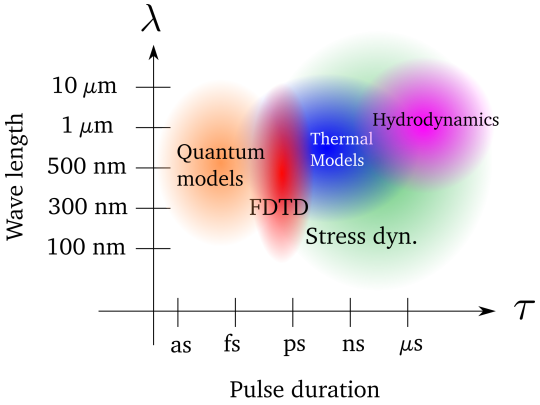
"ATLANTIC": Advanced theoretical network for modeling light matter interaction

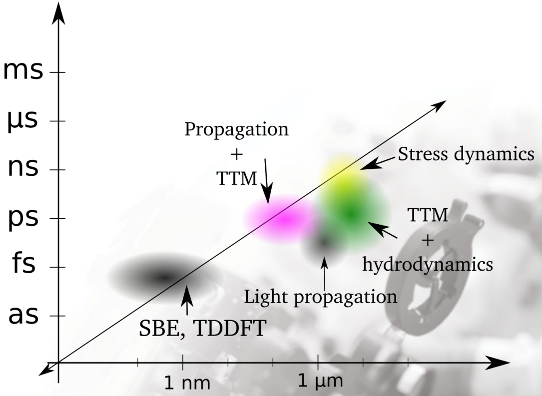


List of theoretical descriptions for laser-matter interaction that are available within the consortium of ATLANTIC project

https://www.QuantumLap.eu/?s=ATLANTIC

Validity ranges of formalisms?





Adapted from proposal H2020-MSCA-RISE-2018 ”ATLANTIC”

Portfolio of Domains and Methods

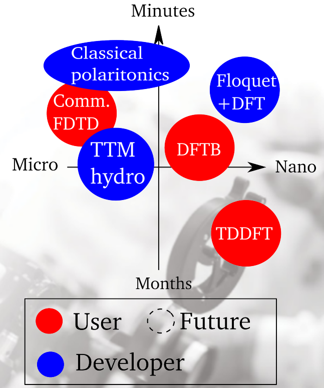
Classical methods ∼ 28+ publications

Domains ↓	Application field ↓\ Methods →	*Thermodyn.* Adv. two-temp. (TTM)	*Thermodyn.* Fluid dyn.	*Class. electrodyn.* Polaritonics	*Class. electrodyn.* FDTD (comm.)
Laser processing	Laser nanostructuring	[1, 5–17]	[18]	[1, 13, 19–22]	[23, 24]
	Damage thresh. prediction	[15, 16, 25–28]	*		
	Film transfer (LIFT)	[28]*			
	Thin film damage/dynamics	[11, 15, 27, 28]	*	[21, 22]	
Materials science	Bulk materials eq. properties			[29]	
	Nanomaterials properties			[22]	[23, 24]
	Photovoltaics				[30]
Ultrafast phenom.	Trans. opt. prop. / metallization	[9–12, 31]		[22]	
	Electron exc. in solids	[1, 9, 16, 25]			
	HHG				
	Decoherence | Collisions	[31]			
	2D materials	[15]			

Quantum methods ∼ 6+ publications

Domains ↓	Application field ↓\ Methods →	Cost → Quantum electrodyn. Floquet+DFT	DFTB	DFT	TDDFT
Laser processing	Laser nanostructuring			[1]	
	Damage thresh. prediction	*			
Materials science	Bulk materials eq. properties			[2]	
	Organic chemistry		[3]		
Ultrafast phenom.	Trans. opt. prop. / metallization	[2]		[2]	[1, 2]
	Electron exc. in solids	[2]		[2]	[2, 4–6]
	HHG			[4, 5]	[1, 4, 6]
	Decoherence | Collisions				[1, 4–6]

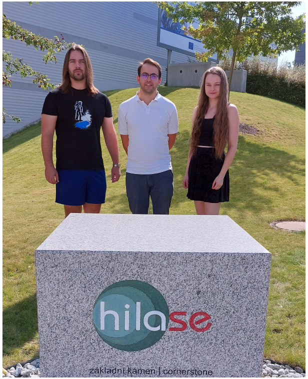
Estimation time vs size



2021-2024: Group ”Ultrafast Photonics” @ HiLASE Centre

Mission ”The [...] group uses condensed matter, quantum formalisms and high-performance computing to invent applications based on phenomena that are induced by ultrashort laser pulses in solids and nanomaterials.”

http://www.QuantumLaP.eu/



2 members

PhD std. Krystof HLINOMAZ

- **classical** thin film thermodynamics,
- **classical** Lagrangian hydro-dynamics.

PhD std. Kristyna GAZDOVA

- Trainee in **quantum** simulations (DFT, Floquet, quantum chem.)

Visiting students (Marie Curie)

PhD std. Andres BERTONI (MSCA-RISE ”ATLANTIC” 4 months, Argentina).

- Transient optical response of solids, **quantum** DFTB

PhD std. Micaela SOSA (MSCA-RISE ”ATLANTIC 2 months, Argentina).

- Transient optical response of **biosystems**

EUROPEAN UNION
European Structural and Investment Funds
Operational Programme Research, Development and Education

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 739573 (HiLASE CoE)

BIATRI
MINISTRY OF EDUCATION, YOUTH AND SPORTS
Science & Technology Facilities Council

2024: Group @ IT4I: ”Quantum Dynamics of Systems”

Laboratory of Quantum Computing (Head: prof. M. Lampart)

Group name: ”Quantum Dynamics of Systems”

People

- PhD appl. *Michal Belina*: quantum chemistry, *ab-initio* molecular dynamics, quantum computing.
- MSc. *Silvie Illésová*: *ab-initio* molecular dynamics, quantum computing.

Scope

- *Ab-initio* dynamics
- Qubit design
- Quantum implementation

PhD appl. Michal Belina

BSc. Silvie Illésová

Outline

1. Context
   - Applications of ultrafast laser-induced phenomena in solids
   - Optical absorption driven by electrons dynamics in solids
     - *Frozen* band structure
     - *Dynamical* band structures: laser dressing was included in Keldysh (1965)
2. TDDFT: multi-band description using high-power computations
   - Modeling the laser excitation of electrons in Si (real-space, real-time TDDFT)
   - Scanning multiple parameters: database preparation
   - Results
3. How reliable are TDDFT predictions in the ultrafast regime? Benchmark vs high harmonic generation (HHG) experiments
4. Predictions of TDDFT at high intensity (laser processing)
   - TDDFT predictions for laser processing: anisotropy in energy absorption & damage threshold
5. Reversible and ultrafast band structure engineering
   - Simplified model: Floquet + DFT
   - Preparation of dipolar matrix elements (DFT)
   - Si [227], LDA: $E_g^\Gamma = 2.56$ eV. Dressing along $K - \Gamma - X$
   - Si [227], LDA: $E_g^\Gamma = 2.56$ eV. "3D" dressed band structure
   - Discussion
6. Overview: predictive modeling tools for high fields photonics are here and they work
7. IT4I/LQC: milestones towards designing a light-controlled multiqubit in solids?

Outline

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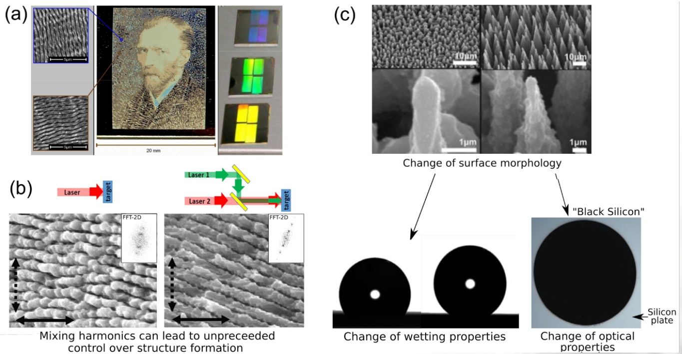
7. IT4I/LQC: milestones towards designing a light-controlled multiqubit in solids?

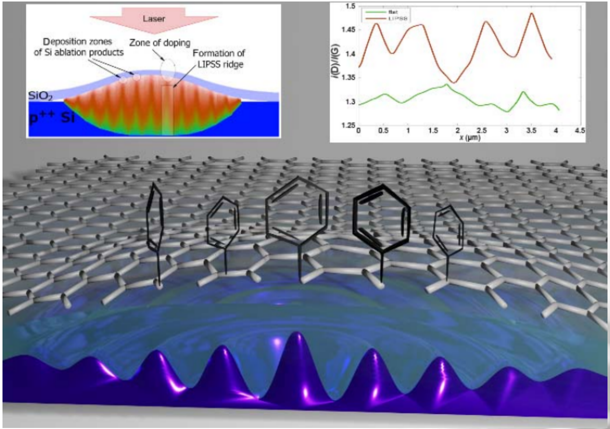
Surface functionalization with intense lasers: the role of polarization



Functionalization of surfaces via laser nanostructuring (source: ”QuantumLaP” MSCA project)

(a) Dusser et al, Opt. Express **18**, 3 (2010)
(b) Jia et al, Phys. Rev. B **72**, 12 (2005)
(c) A. Ranella et al., Acta Biomat. **6**, 2711 (2005)
(d) A.Y. Vorobyev, Ch. Guo, Laser Photon. Rev. **7**, 385 (2013)

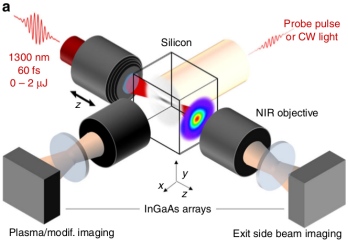
Laser-induced periodic functionalization of graphene

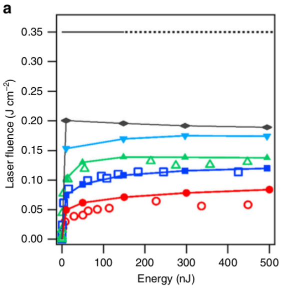


Drogowska-Horna, K. A.; Mirza, I.; [...] Kovaricek P.; [...]; Derrien, T. J.-Y.; [...] Bulgakova, N. M. & Kalbac, M. *Nano Research* (Springer) **13**, 2332 (2020)

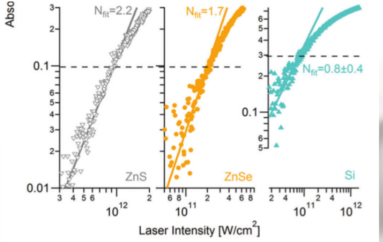
Femtosecond laser modification of bulk crystals: saturation of absorbed energy

Chanal, Grojo et al., *Nature Communications* **8**, 773 (2017). $\lambda = 1300$ nm, $\tau = 60$ fs





Grojo, D.; Utéza, O. et al., *Physical Review B* **88**, 195135 (2013)



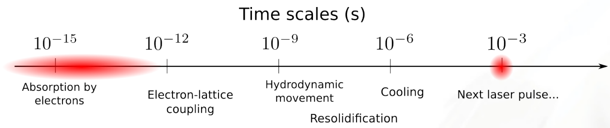
Intensity regime: 10^{12-14} W/cm². Origin of absorption limit?

Macroscopic: defocusing (+ Kerr effect)
Microscopic: increase of the band gap upon excitation? Pauli blocking (saturation of conduction states)?

Physical scales in light-matter interaction

UV - Wavelengths λ - mid-IR

$1\text{ fs} \lesssim$ Pulse durations $\tau \lesssim 20\text{ ps}$



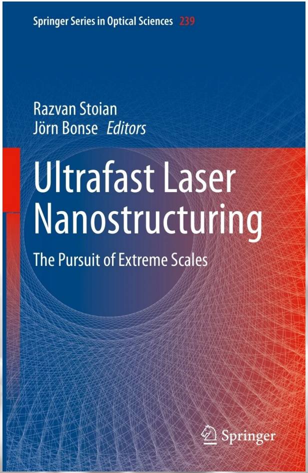
Laser intensity scale

perturbative regime	**material's modification regime**	strong field
$< 10^{11}$ W/cm²	**$10^{11} - 10^{13}$ W/cm²**	10^{14+} W/cm²

Springer book ”The pursuit of extreme scales”

Derrien, T. J.-Y.; Levy, Y. & Bulgakova, N. M.
Chap. 1/33. *Insights into laser-matter interaction from inside: wealth of processes, multiplicity of mechanisms and possible roadmaps for energy localization.*
Ultrafast Laser Nanostructuring - The Pursuit of Extreme Scales (Vol. I-III), Eds: R. Stoian, J. Bonse.
Springer, 2023.

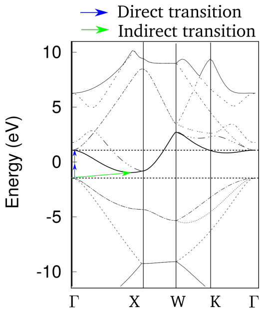
- Vol 1 Fundamentals processes
- Vol 2 Concepts of extreme nanostructuring
- Vol 3 Applications



Outline

Band structure of e- in Si *without* light

To study a material, one usually considers its e- band structure being fixed (*Heisenberg frame*).

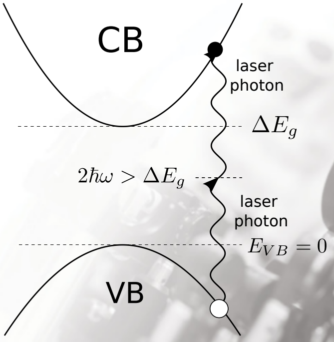


Exp.	Th. (LDA)	Th. (TB09)	Transition	Temperature
3.4 eV	2.56 eV	3.04 eV	$\Gamma \to \Gamma$	0 K
1.16 eV	0.51 eV	0.98 eV	$\Gamma \to X$	0 K

Note: 2.56 eV $\longleftrightarrow$ 484 nm.

Direct or indirect transition?

- Multi-photon absorption is usually direct.

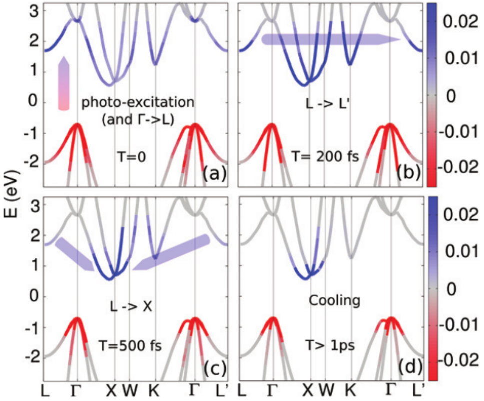


multi-photon absorption

Approximation: fixed electronic bands str. ($\vec{E}$ is $\updownarrow$)

- UV light: 1-photon absorption.
- $I = 10^9$ W/cm^2, $\tau = 110$ fs, $\hbar\omega = 3.4$ eV$= E_{gap}$.

Sangalli et al, *Europhysics Letters* **110**, 47004 (2015)

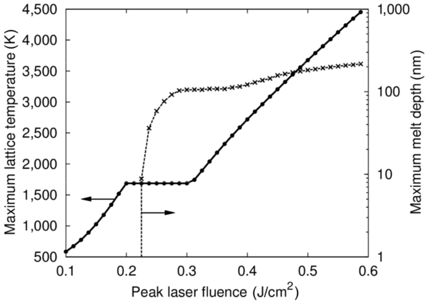


This representation keeps band structure fixed ("Heisenberg frame").
(a) Non-trivial interband transitions.
(b) Intraband transitions: excited electrons transfers between bands.
(c) Relaxation to lower energy levels.

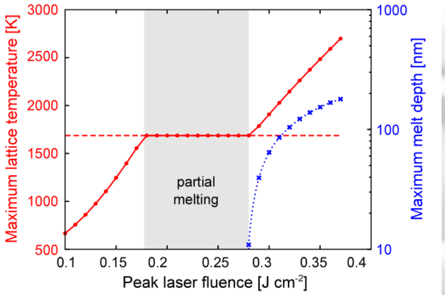
Prediction of melting threshold in Si

Drogowska-Horna, K. A.; Mirza, I.; Rodriguez, A.; Kovaříček, P.; Sládek, J.; **Derrien, T. J.-Y.**; Gedvilas, M.; Raćiukaitis, G.; Frank, O.; Bulgakova, N. M. & Kalbác, M.; *Nano Research*, **13**, 2332-2339 (2020).

Sládek, J.; Levy, Y.; **Derrien, T. J.-Y.**; Bryknar, Z. & Bulgakova, N. M., *Applied Surface Science*, **605**, 154664 (2022)



$\tau = 300$ fs, $\lambda = 1030$ nm



$\tau = 250$ fs, $\lambda = 1030$ nm

$\tau_{\text{laser}} > \tau_{\text{e-ph}}$: two-temperature modeling. Pump-probe reflectivity

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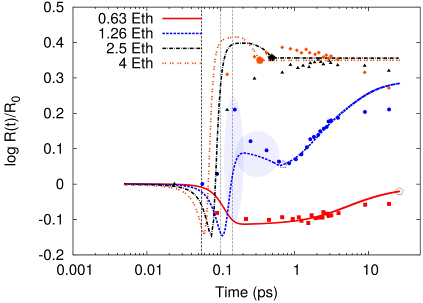
Derrien, T. J.-Y. & Bulgakova, N. M. Proc. SPIE **10228** (2017)



—: our **theory**. □: experimental from Shank, C. et al.,, *Phys. Rev. Lett.*, **50**, 454 (1983).
⬠: partial melting starts, filled-⬠: total melting is achieved (at least one cell). $\tau = 90$ fs, $\lambda_P = 620$ nm, $\lambda_p = 1$ µm.

Advantages of TTM

- Pump-probe reflectivity as function of time and energy.
- Spatial-dependence & energy transport.
- Importance of 3-body phenomena (screening of e-ph coupling, Auger recombination, ...).

Limits of *two-temperature model* for band-gap materials

- Free param. (σ_1, σ_2, ν, m^*, ...) → fitting procedure → "predictions"
- Excessive material dependency. Dependent on crystal orientation.
- Limited to $\tau > \tau_{e-ph}$, $\tau > \tau_{e-e}$

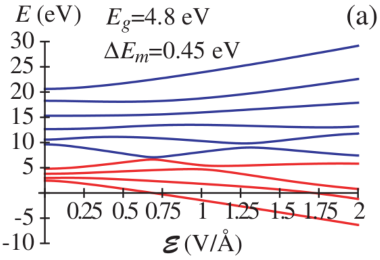
Predicting → no parameter fitting → 1st principles

Necessity to get rid of free-parameters

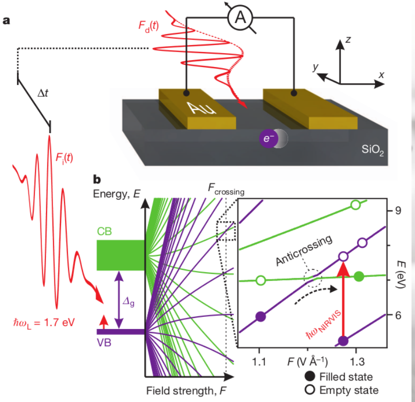
2011: ($\tau < \tau_{eph}$) reversible metallization of dielectrics ($\vec{E}$ is $\updownarrow$)

2011 Durach, M.; Rusina, A.; Kling, M. F. & Stockman, M. I. Predicted **Ultrafast Dynamic Metallization of Dielectric Nanofilms** by Strong Single-Cycle Optical Fields, *Phys. Rev. Lett.* **107**, 086602 (2011)

2012 Schiffrin, A. [...]; Stockman, M. I. & Krausz, F. et al., Optical-field-induced current in dielectrics, *Nature* **493**, 70 (2012)

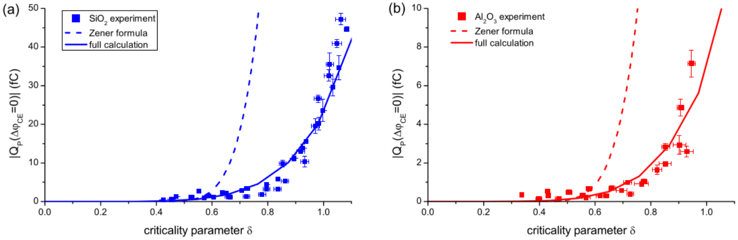


Phys. Rev. Lett. **107**, 086602 (2011)



Laser-assisted PHz diode: Zenner tunneling ($\vec{E}$ is $\updownarrow$)

2016 Kwon, O.; [...] Kim, B.-K.; Kim, J.-J.; Stockman, M. I. & Kim, D. Semimetallization of dielectrics in strong optical fields, *Scientific Reports*, **6**, 21272 (2016)



Q_p/[Coulomb]: transfered charges per pulse.
δ/ [V/m]: laser field strengh

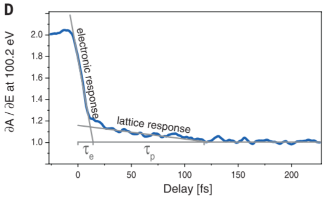
”Universal” ultrafast phenomenon? Yes & No.

Field-assisted metallization demonstrated for SiO_2, Al_2O_3, and BaF_2.

"Ultrafast" idea: act *before* phonons kick in

Role of phonons

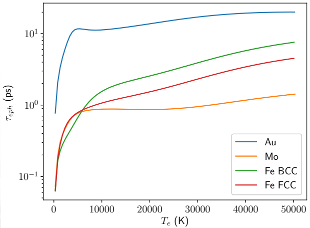
- $\tau, t < \gamma_{\text{e-ph}}^{-1}$: assuming an "optical" regime;
- $\tau, t > \gamma_{\text{e-ph}}^{-1}$: assuming a thermodynamical "collisional" regime: $\nu = f(T_e, T_{\text{lattice}})$.



Si: Schultze, M. & al. Science **346**, 1348-1352 (2014)

Material	e-ph coupling time	Ref.
Si	$\gtrsim$240 fs (exp.)	Sjodin et al. PRL (1998)
Si	$\sim$64 fs (exp.)	Schultze et al. Science (2014)
Au	770 fs - 20 ps (th.)	Lin et al. PRB (2008)
Mo	70 fs - 1.4 ps (th).	Lin et al. PRB (2008)

Electron-phonon coupling times in various crystals.

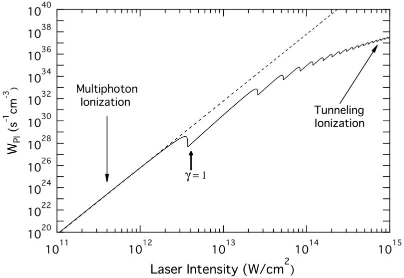


Reconstructed from Lin, Z.; Zhigilei, L. V. & Celli, V. *Physical Review B* **77**, 075133 (2008)

Approximation for our "semi-quantum" works

We consider pulses $\tau \ll \gamma_{\text{e-ph}}^{-1}$, and disregard effect of lattice $\rightarrow$ **direct** transitions at Γ.

Laser effect on optical band gap?



Gulley, J. R., *Opt. Eng.* **51**, 121805-1 (2012)

Band gap dynamics in Keldysh (1965) vs reality ($\vec{E}$↕)?

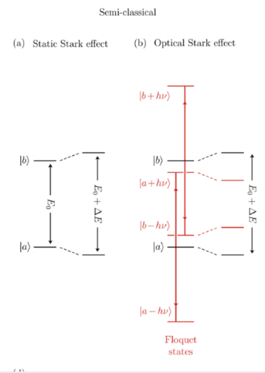
- **In MPI regime**, effective band gap ↑ with laser field [Gulley, J. R. Opt. Eng. **51**, 121805-1 (2012)].

$$E_g^{\text{eff}}(F) \underset{\gamma \gg 1}{\simeq} E_g + \frac{e^2 F^2}{4 m_e \omega^2} \text{ (Kane).}$$

- **Tunneling** rather ↓ effective band gap energy E_g^{eff} with intensity.
- Is number of photons required for e- transition ↑ or ↓ with laser intensity?

Laser dressing: optical Stark effect ($\vec{E}$is ↻)

Sie, E. [...] Gedik, N. et al., Valley-selective optical Stark effect in monolayer WS_2, *Nature Materials*, **14**, 290-294 (2014)



Dressing is included in Keldysh model ($\vec{E}\updownarrow$)

VSB TUO | IT4I

Popular aspects

Adiabadicity parameter: $\gamma \hat{=} \frac{t_{\text{tunneling}}}{T_{\text{laser}}}$.
$\gamma \ll 1$: "tunneling dominates",
$\gamma \gg 1$: "multiphotonic transitions dominate."

Hamiltonian expressed in the *length gauge and dipolar approximation* [Keldysh, L. Behavior of non-metallic crystals in strong electric fields, *Sov. Phys. JETP* **6**, 763 (1958)]:

$$\hat{H} = \hat{H}_{0e} + \hat{H}_{0L} + \hat{H}_{eL} + e\mathbf{E}\mathbf{r};$$

$$\hat{H}_{0e} = \frac{1}{2m}\left(\frac{\hbar}{i}\nabla\right)^2 + W(\mathbf{r});\ \hat{H}_{0e}\psi_{0j}(\mathbf{p},\mathbf{r}) = \varepsilon_j(\mathbf{p})\psi_{0j}(\mathbf{p},\mathbf{r});$$

Keldysh, L. Sov. Phys. JETP **47**, 1307-1314 (1964).

1. Ionization **probability** obtained from *Fermi golden rule*, using *Houston wave functions*.

$$H_0(t) = \int \psi_0^+(\mathbf{r}t)\left\{\varepsilon\left(-i\hbar\nabla - \frac{e}{c}\mathbf{A}\right) + e\Phi\right\}$$

$$\times\ \psi_0(\mathbf{r}t)\,d\mathbf{r} + H_T. \qquad (3)$$

The Bloch wave functions of an electron, accelerated by the field inside each of the bands, have a form analogous to (6):

$$\psi_{\mathbf{p}}^{c,v}(\mathbf{r},t) = u_{\mathbf{p}(t)}^{c,v}(\mathbf{r})\exp\left\{\frac{i}{\hbar}\left[\mathbf{p}(t)\mathbf{r} - \int_0^t \varepsilon_{c,v}(\mathbf{p}(\tau))\,d\tau\right]\right\},$$

$$\mathbf{p}(t) = \mathbf{p} + (e\mathbf{F}/\omega)\sin\omega t, \qquad (26)$$

where $u_{\mathbf{p}}^{c,v}(\mathbf{r})$ are periodic functions that have the translational symmetry of the lattice. Calculations perfectly similar to (8)—(15) lead to a general formula for the ionization probability

$$w = \frac{2\pi}{\hbar}\int\frac{d^3p}{(2\pi\hbar)^3}\,|L_{cv}(\mathbf{p})|^2\sum_n \delta(\overline{\varepsilon(\mathbf{p})} - n\hbar\omega), \qquad (27)$$

2. **Dressing** of 2 electronic levels by the photon field.

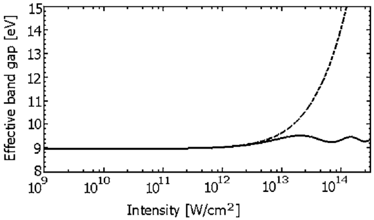
$$\overline{\varepsilon(\mathbf{p})} = \frac{1}{2\pi}\int_{-\pi}^{\pi}\varepsilon\left(\mathbf{p} + \frac{e\mathbf{F}}{\omega}\sin x\right)dx,$$

A simplified dressing is included in Keldysh theories

2-bands description of dressing of electronic levels.
In MPI regime, effective band gap **increases** with field strength. In tunneling, it should "**decrease**."

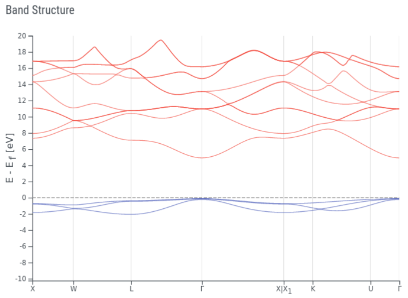
Band gap dynamics in the laser field $E_{gap}[\varepsilon(\mathrm{k}+\mathrm{A})]$?

Gruzdev, V. Photoionization rate in wide band-gap crystals. *Physical Review B*, **75**, 205106 (2007).



—: cosine dispersion, ···: Kane dispersion (NaCl)

Approached band structure of NaCl (225) crystal (topological materials.org)



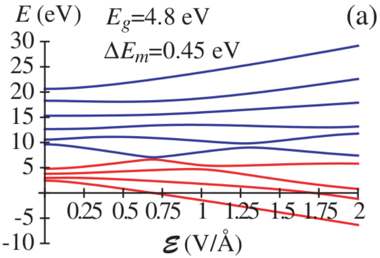
Limits of the 2-band approximation

- Conditions for band gap opening with field?
- Is dispersion law sufficient to estimate a dynamical band gap variation?
- What happens if we account for more bands?

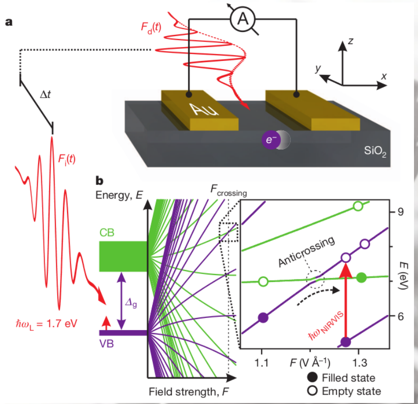
Reversible metallization of dielectrics

2011 Durach, M.; Rusina, A.; Kling, M. F. & Stockman, M. I. Predicted **Ultrafast Dynamic Metallization of Dielectric Nanofilms** by Strong Single-Cycle Optical Fields, *Phys. Rev. Lett.* **107**, 086602 (2011)

2012 Schiffrin, A. [...]; Stockman, M. I. & Krausz, F. et al., Optical-field-induced current in dielectrics, *Nature* **493**, 70 (2012)



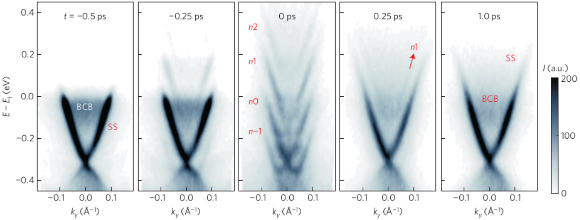
Phys. Rev. Lett. **107**, 086602 (2011)



Transient states in Bi_2Se_3 ($\lesssim$77 K, $\vec{E}$: ↕ vs ↻)

2016 Mahmood, F.; Chan, C.-K.; Alpichshev, Z.; Gardner, D.; Lee, Y.; Lee, P. A. & Gedik, N. *Nature Physics*, **12**, 306-310 (2016)

2013 Wang, Y. H.; Steinberg, H.; Jarillo-Herrero, P. & Gedik, N. *Science* **342**, 453-457 (2013)



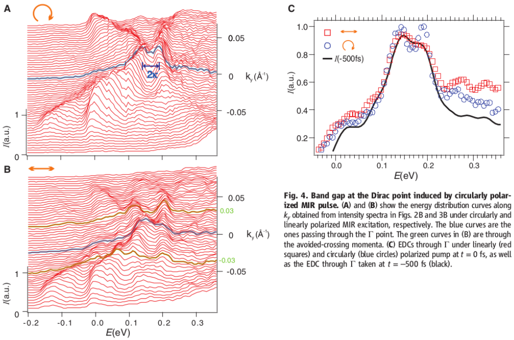


Fig. 4. Band gap at the Dirac point induced by circularly polarized MIR pulse. (**A**) and (**B**) show the energy distribution curves along k_y obtained from intensity spectra in Figs. 2B and 3B under circularly and linearly polarized MIR excitation, respectively. The blue curves are the ones passing through the Γ point. The green curves in (B) are through the avoided-crossing momenta. (**C**) EDCs through Γ under linearly (red squares) and circularly (blue circles) polarized pump at t = 0 fs, as well as the EDC through Γ taken at t = −500 fs (black).

Linear polarization ($\vec{E}$ ↕): replication of band structure

- Transient Wannier-Stark ladder observed in solids (i.e., dressed picture is meaningful).
- Accessible via TD-ARPES & AR-pump-probe.
- Band gap closes during the pulse.

Circular polarization ($\vec{E}$ ↻): potential opening of gap

Only in two-dimensional & topological materials.

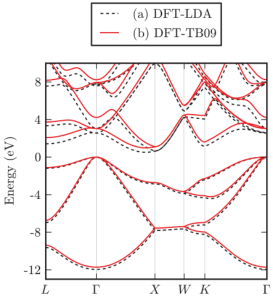
Outline

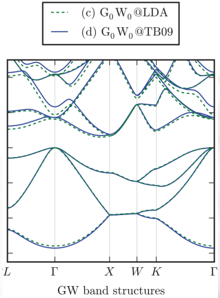
Outline

1. Context
   - Applications of ultrafast laser-induced phenomena in solids
   - Optical absorption driven by electrons dynamics in solids
     - *Frozen* band structure
     - *Dynamical* band structures: laser dressing was included in Keldysh (1965)
2. TDDFT: multi-band description using high-power computations
   - Modeling the laser excitation of electrons in Si (real-space, real-time TDDFT)
   - Scanning multiple parameters: database preparation
   - Results
3. How reliable are TDDFT predictions in the ultrafast regime? Benchmark vs high harmonic generation (HHG) experiments
4. Predictions of TDDFT at high intensity (laser processing)
   - TDDFT predictions for laser processing: anisotropy in energy absorption & damage threshold
5. Reversible and ultrafast band structure engineering
   - Simplified model: Floquet + DFT
   - Preparation of dipolar matrix elements (DFT)
   - Si [227], LDA: $E_g^\Gamma = 2.56$ eV. Dressing along $K - \Gamma - X$
   - Si [227], LDA: $E_g^\Gamma = 2.56$ eV. "3D" dressed band structure
   - Discussion
6. Overview: predictive modeling tools for high fields photonics are here and they work
7. IT4I/LQC: milestones towards designing a light-controlled multiqubit in solids?

Band structure at 0K

Waroquiers, D.; Lherbier, A.; Miglio, A.; Stankovski, M.; Poncé, S.; Oliveira, M. J. T.; Giantomassi, M.; Rignanese, G.-M. & Gonze, X. *Physical Review B*, **87**, 075121 (2013)





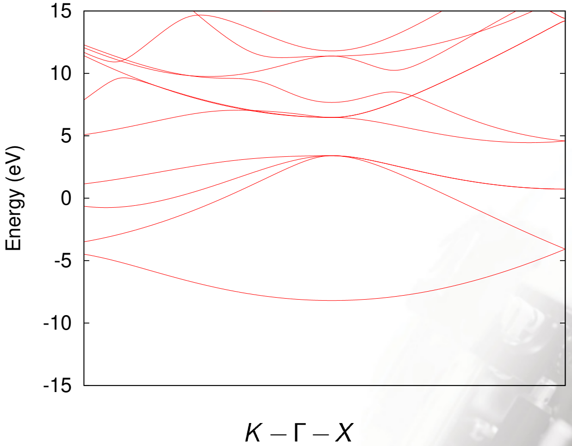
DFT band gaps @ 0K (dir. and indir.)

DFT+LDA: 2.56 eV and 0.51 eV.
DFT+TB09: 3.04 eV and 0.98 eV.

GW band gaps @ 0K (dir. and indir.)

GW+LDA: 3.25 eV and 1.21 eV.
GW+TB09: 3.44 eV and 1.38 eV.

Band structure TB09 (T=0 K)



Direct band gap energy: 3.06 eV. Indirect band gap enregy: 1.04 eV.

TDDFT simulations

TDDFT: "time-dependent **density** functional theory"

Kohn-Sham equation in a solid using the method of *ab-initio* norm-conserving pseudo-potentials

$$\left[\left(\underbrace{-\frac{i\hbar}{2m_e}\nabla_{\mathrm{r}}}_{\text{kinetic energy}}+\frac{|e|}{c}\mathrm{A}(t)\right)^2+\underbrace{\hat{v}_{\mathrm{ion}}(\mathrm{r})}_{\text{atoms}}+\underbrace{\hat{v}_{\mathrm{H}}[n(\mathrm{r},t)]}_{\text{e- density-functional}}(\mathrm{r})+\right. \tag{1}$$

$$\left.+\underbrace{\hat{v}_{\mathrm{xc}}[n(\mathrm{r},t)]}_{\text{e- density-functional}}(\mathrm{r})\right]\times\psi_{n,\mathrm{k}}(\mathrm{r},t)=i\hbar\frac{\partial}{\partial t}\psi_{n,\mathrm{k}}(\mathrm{r},t)$$

with vector potential (dipolar approximation)

$$\mathrm{A}(t)=-c\int_{-\infty}^{t}\underbrace{\mathrm{E}(t')}_{\text{laser light}}\,dt', \tag{2}$$

expresssed in atomic units (Hartree).

Some of ”Top500” computers used in our works

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”Salomon” and ”Barbora”, IT4I, Ostrava, Czech Republic

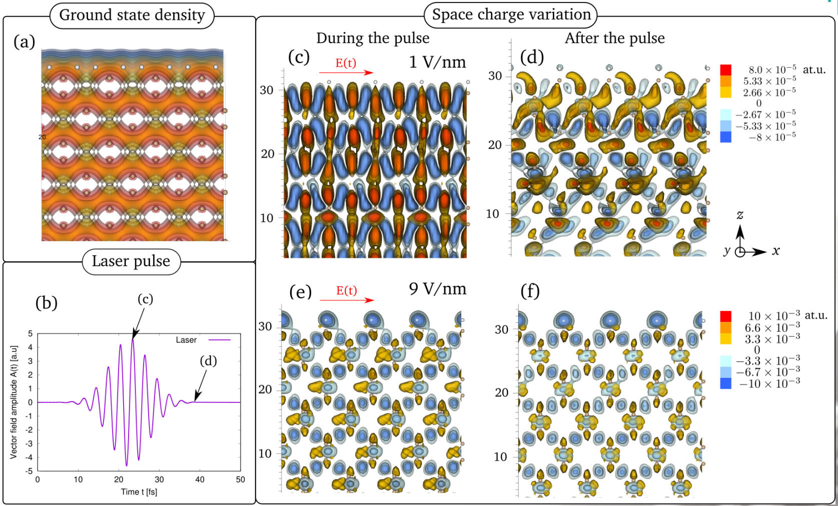
”Karolina”, IT4I, Ostrava, Czech Republic

Experience with Top500 supercomputers

Top500.org: world-chart of the most efficient computers in the world.

Era	Machine	Institute	Top500	Country	System
2021+	*Karolina CPU*	*IT4I*	*149*	*Czech Republic*	*Linux*
2016-2019	Draco	Max Planck	160	Germany	Linux
2016	EOS	Max Planck	264	Germany	Linux
2015-2020	Salomon	IT4I	40	Czech Republic	Linux
2019-2020	Prometheus	Cyfronet	49	Poland	Linux
2010	Jade	Cines.fr	18	France	Linux

Ex: charge density fluctuations



Derrien, T. J.-Y.; Tancogne-Dejean, N.; Zhukov, V.; Appel, H.; Rubio, A. & Bulgakova, N. M. Unpublished material.

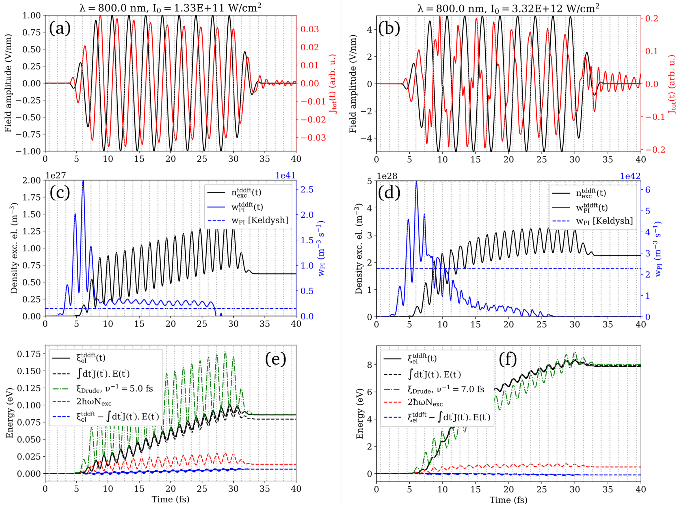
Si slab: real-space visualization of electron oscillation

Video: click on the screen.

Si bulk: real-space visualization of electron oscillation

Video: click on the screen.

Ex: $n_{\text{exc}}(t)$, $J(t)$, energy $\xi_{e^-}(t)$



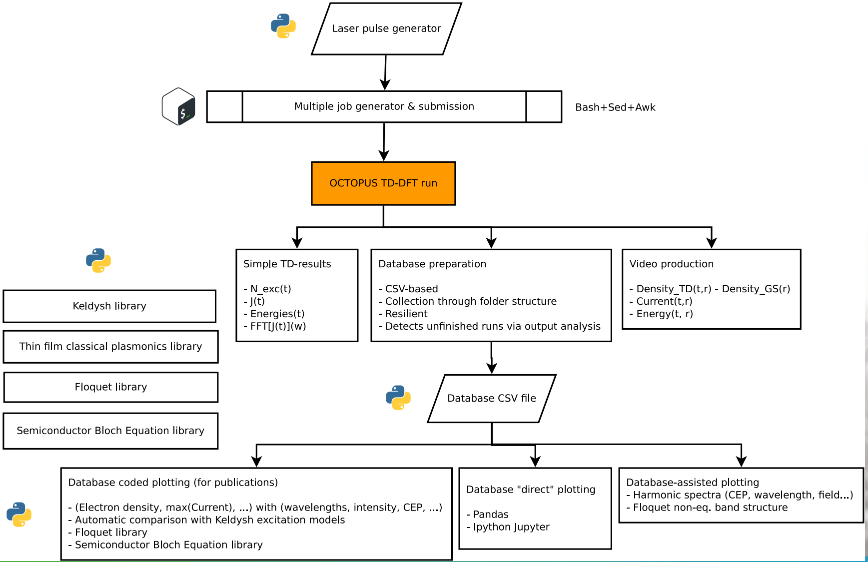
Left: $E = 1$ V/nm. Right: $E = 5$ V/nm.

Derrien, T. J.-Y.; Tancogne-Dejean, N.; Zhukov, V.; Appel, H.; Rubio, A. & Bulgakova, N. M.

Outline

In-house library: ”octopus-slabs”

Product: In-house library ”octopus-slabs”.



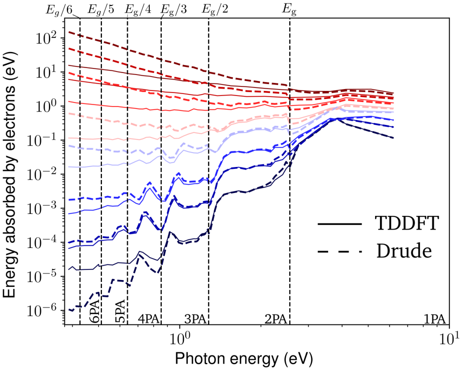
Code development

Since 2016 Multi-year investment @ 1 FTE.
2016-2023 Hosted by *BitBucket.org* (academic license 0€)
Since 2023 Hosted by *GitHub.com* (PRIVATE REPO).
https://github.com/tjyderrien/octopus-slabs/

Outline

Absorbed energy: TDDFT vs Drude model

Derrien, T. J.-Y.; Tancogne-Dejean, N.; Zhukov, V.; Appel, H.; Rubio, A. & Bulgakova, N. M.
Phys. Rev. B, **104** L241201 (2021)



Observation

- Prediction of the absorbed electron energy from TDDFT. Multiphotonic peaks are clearly visible.
- 1 Drude model applied to all wavelengths and intensities using 1 collision frequency $\nu^{-1} = 6$ fs (indep. from field or wavelength).

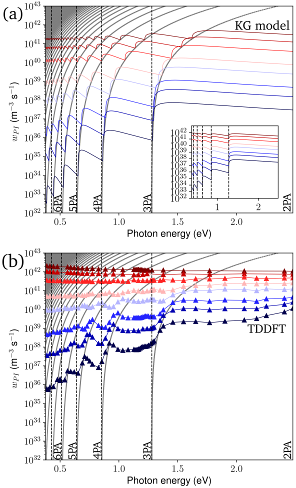
Questions

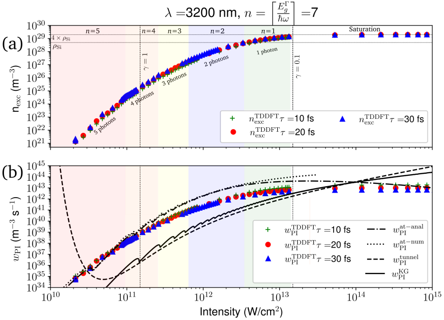
- How accurate are TDDFT results vs experiments? In which regime?
- Could damage threshold of e.g. Si ($\tau < \tau_{eph}$) be studied by combining Keldysh & Drude models inside a thermal model?
- Applications of the transient band gap dynamics?

[1] **Product**: prediction of excitation rates and absorbed energy for laser processing labs/companies...

Excitation: TDDFT vs Keldysh model for Si ($\vec{E}$ is $\updownarrow$)

Derrien, T. J.-Y.; Tancogne-Dejean, N.; Zhukov, V.; Appel, H.; Rubio, A. & Bulgakova, N. M. *Phys. Rev. B*, **104** L241201 (2021)





Keldysh model (1964): limited qualitative agreement w/ TDDFT

- **Keldysh excitation rate w_{PI}**: *qualitative* agreement for Si. Agreement as function of wavelength was impressive (Si: $\Gamma \to \Gamma$, valid for $\tau < 30$ fs).
- **Band-gap decreases** with intensity: light-induced tunneling. 5 ph. $\to 4 \to 3 \to 2 \to 1 \to 0$.

Example of TDDFT-generated excitation rates

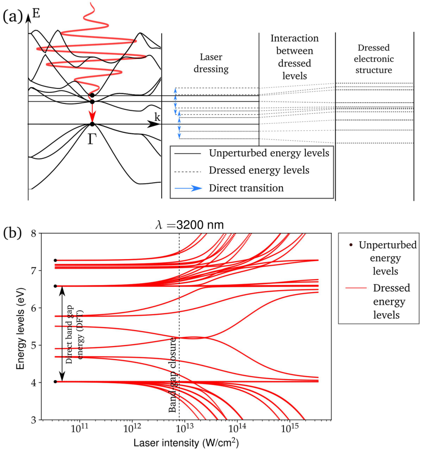
Derrien, T. J.-Y.; Tancogne-Dejean, N.; Zhukov, V.; Appel, H.; Rubio, A. & Bulgakova, N. M.
Phys. Rev. B, **104** L241201 (2021)

Method	Wavelength	τ_p	Band gap	Intensity range (W/cm^2)	Eff. transition probability	Ref.
Theory (TD-LDA)	3200 nm	30 fs	2.56 eV (d)	$(2.1-9.9)\times10^{10}$	$\sigma_5\ (\mathrm{m^7W^{-4}}) = 4.84\times10^{-56}$	This work
				$(1.0-2.6)\times10^{11}$	$\sigma_4\ (\mathrm{m^5W^{-3}}) = 3.05\times10^{-41}$	This work
				$(2.6-5.3)\times10^{11}$	$\sigma_3\ (\mathrm{m^3W^{-2}}) = 5.25\times10^{-26}$	This work
				$(0.53-3.4)\times10^{12}$	$\sigma_2\ (\mathrm{mW^{-1}}) = 2.00\times10^{-10}$	This work
				$(0.34-1.0)\times10^{13}$	$\sigma_1\ (\mathrm{m^{-1}}) = 2.93\times10^{6}$	This work
Exp.		200 fs			$\sigma_3\ (\mathrm{m^3W^{-2}}) = 0.5\times10^{-26}$	Pearl et al. (2008)

More is available for direct transitions

- See Suppl. Inf. of the paper. [Derrien *et al.*, PRB **104** L241201 (2021)].
- TDDFT database at `QuantumLaP.eu`

Ultrafast band-gap closure in semi-conductors



T. J.-Y. Derrien, N. Tancogne-Dejean, [...] and N. M. Bulgakova, *Phys. Rev. B*. **104** L241201 (2021)

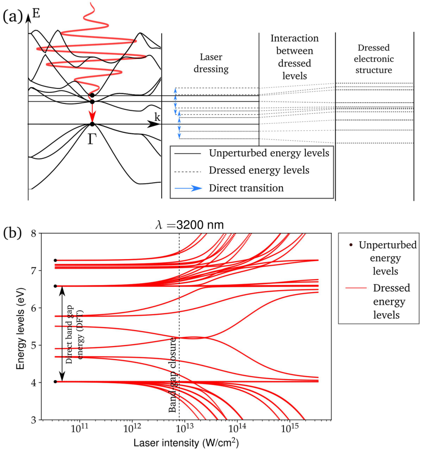
Effect of the laser excitation ($\vec{E}\updownarrow$) on the band structure?

Upon fs irradiation, ”trivial” band-gap materials become metallic above a threshold intensity.

Joint work with Kristyna Gazdova & Andrés I. Bertoni

$\lambda = 1030$ nm, $E \leq 0.05$ V/nm (~3×10^{10} W/cm^2).

Ultrafast band-gap closure in semi-conductors



T. J.-Y. Derrien, N. Tancogne-Dejean, [...] and N. M. Bulgakova, *Phys. Rev. B.* **104** L241201 (2021)

Effect of the laser excitation ($\vec{E}$ ↕) on the band structure?

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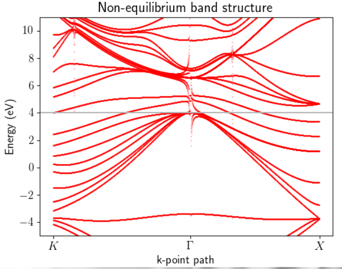

$\lambda = 1030$ nm, $E \leq 0.05$ V/nm (~3×10^{10} W/cm^2).

More: database of excitation rates, absorbed energy, ...

For Si, +3,000 TDDFT simulations with relevant laser pulses have been prepared
[~M-core-hours per year]

- Several materials (Si, SiO_2, Mo, Au, ...)
- Several pulse shapes, pulse mixtures, ...
- Several observables (absorbed energy, currents, harmonic spectra, ...).
- All the work has been systematized into PYTHON & BASH routines for collaboration purposes.

High Power Computation Projects

- **IT4Innovations** National Supercomputing Center - eINFRA (ID:90140), sub-proj. MORILLE, FLAMENCO, FILIPINAS.
- **PRACE** aisbl (projects BOLERO, FRECUENCIA).

Backup National Grid Infrastructure **MetaCentrum** eINFRA (ID:90140).

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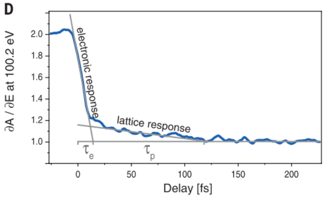
Outline

1. Context
2. TDDFT: multi-band description using high-power computations
3. **How reliable are TDDFT predictions in the ultrafast regime? Benchmark vs high harmonic generation (HHG) experiments**
4. Predictions of TDDFT at high intensity (laser processing)
5. Reversible and ultrafast band structure engineering
6. Overview: predictive modeling tools for high fields photonics are here and they work
7. IT4I/LQC: milestones towards designing a light-controlled multiqubit in solids?

"Ultrafast" regime: disregarding phonons.

Role of phonons

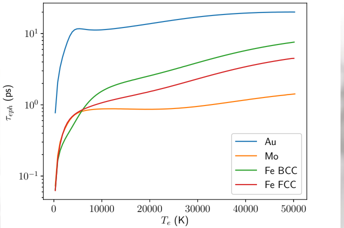
- $\tau, t < \gamma_{\text{e-ph}}^{-1}$: assuming an "optical" regime;
- $\tau, t > \gamma_{\text{e-ph}}^{-1}$: assuming a thermodynamical "collisional" regime: $\nu = f(T_e, T_{\text{lattice}})$.



Si: Schultze, M. & al. Science **346**, 1348-1352 (2014)

Material	e-ph coupling time	Ref.
Si	$\gtrsim$240 fs (exp.)	Sjodin et al. PRL (1998)
Si	$\sim$64 fs (exp.)	Schultze et al. Science (2014)
Au	**770** fs - 20 ps (th.)	Lin et al. PRB (2008)
Mo	**70** fs - **1.4** ps (th).	Lin et al. PRB (2008)

Electron-phonon coupling times in various crystals.



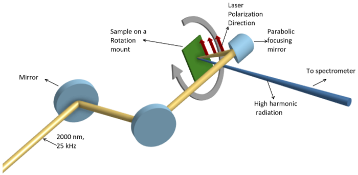
Reconstructed from Lin, Z.; Zhigilei, L. V. & Celli, V. *Physical Review B* **77**, 075133 (2008)

Choice for this work

We consider pulses $\tau \ll \gamma_{\text{e-ph}}^{-1}$, and disregard effect of lattice $\rightarrow$ direct transitions at Γ.

HHG experiments in reflection @ Charles Uni., Prague

- Group of assoc. prof. Martin Kozak.
- $\tau = 15$ fs or 25 fs at FWHM.
- Wavelength $\lambda \sim 2000$ nm.
- Electric field: $E = 3$ V/nm out of sample.
- Probing harmonics generated in reflection configuration.



Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & **Kozák, M.**, *Comm. Phys.* **5**, 288 (2022).

"Ultrashort" pulses

Experimental

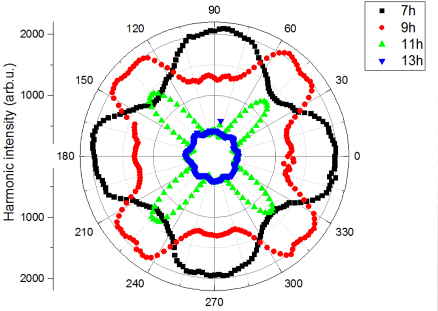
Quantity	Notation	Value	Unit
Pulse duration	τ	25	fs
Wavelength	λ	2000	nm
Band gap energy	E_g^Γ	3.4	eV
Optical cycles	n_{OC}	3.747	cycles

Table: 25 fs FWHM pulses

Experiment: HHG(ϕ, n)

Dr. Martin Kozak group (Charles University). Experimental measurement of harmonic spectrum emitted by Si as function of sample orientation.

- n: harmonic order.
- ϕ: orientation angle in plane ($[100], [110]$).



Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M., *Comm. Phys.* **5**, 288 (2022).

Observables

Excited electron density (Otobe et al., 2008)

$$n_{\text{exc}}(t) = \frac{1}{V}\left[N_{\text{tot}} - \sum_{n,n',\mathrm{k}}^{\text{occ.}} \left| \int d^3\mathrm{r}\ \underbrace{\psi^{\dagger}_{n',\mathrm{k}}(\mathrm{r},t)}_{\text{time-evolved}}\ \underbrace{\psi^{\text{GS}}_{n,\mathrm{k}}(\mathrm{r})}_{\text{ground state}} \right|^2\right]. \tag{3}$$

Total current (Stefanucci & Leeuwen, 2013)

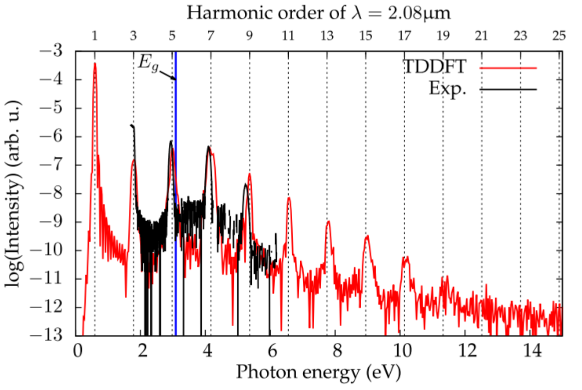
$$\mathrm{J}(t) \propto \left(\nabla\psi^{\dagger}\right)\psi - \psi^{\dagger}\left(\nabla\psi\right)$$

Harmonic spectrum (Larmor formula) (Floss et al., 2018; Tancogne-Dejean et al., 2017)

$$\text{HHG}(\omega) = \omega^2 \left|\mathscr{F}\left[\mathrm{J}(t)\right](\omega)\right|^2.$$

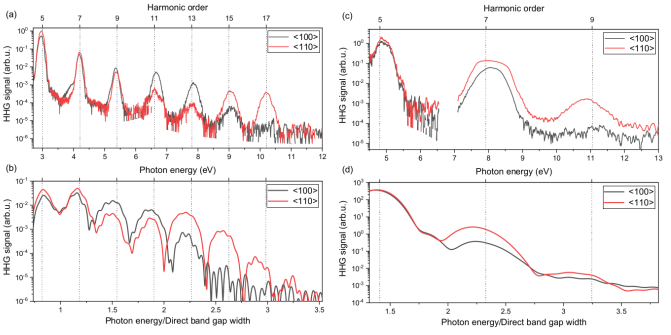
State of art in Germany

Simulation of high-harmonic generation has been historically difficult.



Max Planck Institute: Klemke *et al. Nat. Comm.* **10**, 1319 (2019): $\tau = 50$ fs, $\lambda = 2080$ nm, $\sin^2$, TB09.

CZ: HHG experiment vs TDDFT



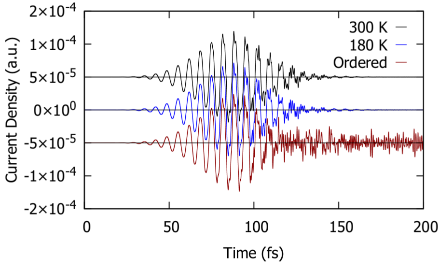
Czech Republic: Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M., *Comm. Phys.* **5**, 288 (2022). [(a,c): exp., (b,d): TDDFT. (a,b): $\lambda = 2000$ nm, (c,d): $\lambda = 800$ nm].

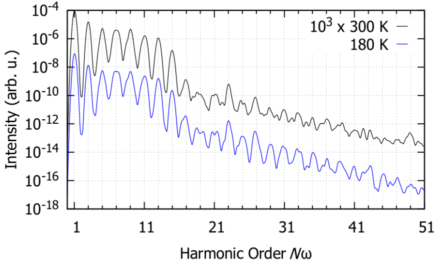
Ultrafast photonic applications can rely on predictions provided by TDDFT

- High quality predictions: model matches with experiments.
- Can be applied to other materials. Can be improved by introducing real shape of pulse.

Effect of temperature on HHG spectra, *ab-initio*

Japan: Freeman, D.; Kheifets, A.; Yamada, S.; Yamada, A. & Yabana, K. High-order harmonic generation in semiconductors driven at near- and mid-infrared wavelengths, *Phys. Rev. B*, **106**, 075202 (2022)



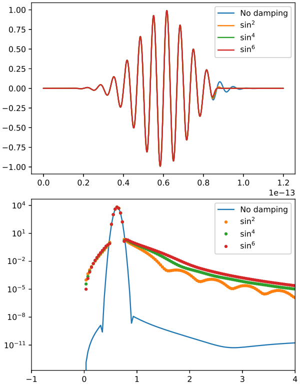


Effect of lattice temperature? Not much in Si

- $J(t)$: temperature $\rightarrow$ a source for damping the oscillations of electrons.
- Harmonic spectrum is sensitive to ultracold environments (space!)

Introducing losses?

In pure TDDFT, $J(t)$ is eternally oscillating (no damping). To avoid inducing a non-physical discontinuity at the end of simulation, damping is applied to currents.



Method for damping the tail of the current $J(t)$

$$J_{\text{damped}}(t) = J(t) \times \eta(t)$$

where

$$\eta(t) = 1 - H(t - t_{\text{final}} + t_{\text{damping}}) + \\ + \sin^2\left[\frac{\pi}{2} \times \frac{t - t_{\text{final}}}{t_{\text{damping}}}\right] \\ \times H(t - t_{\text{final}} + t_{\text{damping}}) \\ \times [1 - H(t - t_{\text{final}})]$$

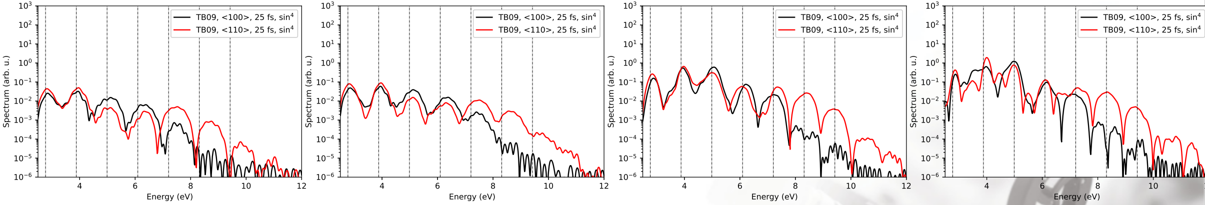
and $H(t)$ is the Heaviside function.

In all the following slides

This damping is applied to all $J(t)$, and impacts HHG(ω) distribution.

Effect of ”decoherence time”

$$\text{HHG}(\phi = 0^\circ, 45^\circ) \text{ - TB09 - } \tau = 25 \text{ fs - } \sin^4$$



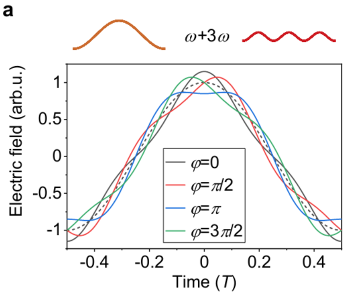
1.25τ 1.3τ 1.5τ 1.75τ

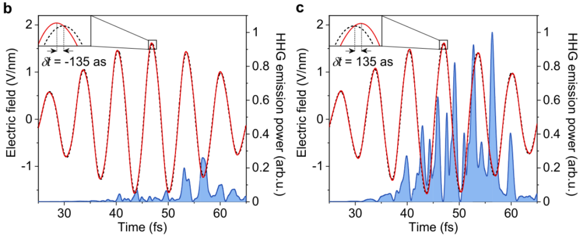
Importance of decoherence & damping

Temporal dynamics is crucial: collisions and decoherence induce discrepency of TDDFT vs TD-experiments.

Latest benchmark: HHG $\omega + 3\omega$ vs TDDFT

Gindl, A.; Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M.
Attosecond control of solid-state high harmonic generation using ω-3ω fields, arXiv:2310.07254
$\tau = 30$ fs, $\lambda_1 = 2000$ nm, $\lambda_2 = 666$ nm. $E \sim 1.5$ V/nm ($I \sim 0.3 \times 10^{12}$ W/cm^2 or 8 mJ/cm^2).





Effect of CEP on harmonic emission (filtered HHG yield)

Our TDDFT predictions reveal the extreme sensitivity of HHG to CEP

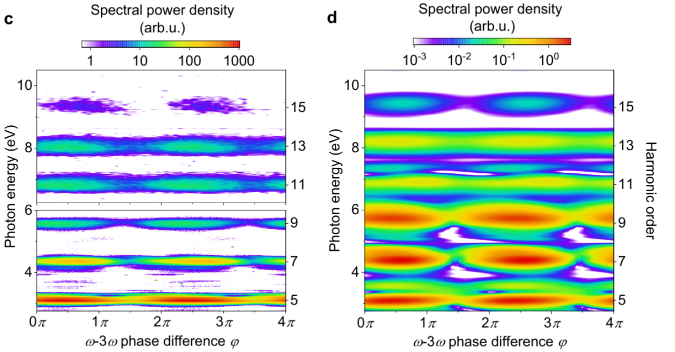
Choice of carrier-envelope phase (CEP) has large influence on attosecond electron excitation dynamics in solids.
→ **Coherent control of electron dynamics** possible in the bi-color mixing regime.

Exp. HHG $\omega + 3\omega$ vs TDDFT predictions

Gindl, A.; Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M.

Attosecond control of solid-state high harmonic generation using ω-3ω fields, arXiv:2310.07254

$\tau = 30$ fs, $\lambda_1 = 2000$ nm, $\lambda_2 = 666$ nm. $E \sim 1.5$ V/nm ($I \sim 0.3 \times 10^{12}$ W/cm^2 or 8 mJ/cm^2).



HHG experiments ← | →: TDDFT simulations

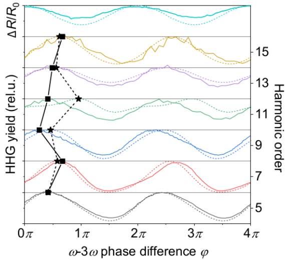
Our TDDFT predictions vs attosecond HHG experiment

Phase-dependent **agreement** for multiple harmonic orders in the **tunneling regime**

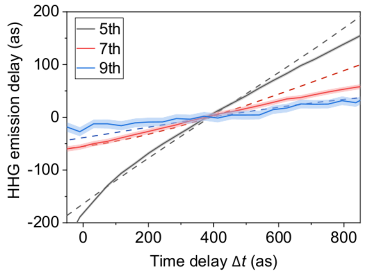
HHG $\omega + 3\omega$ vs TDDFT: high prediction capabilities

Gindl, A.; Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M.
Attosecond control of solid-state high harmonic generation using ω-3ω fields, arXiv:2310.07254
$\tau = 30$ fs, $\lambda_1 = 2000$ nm, $\lambda_2 = 666$ nm. $E \sim 1.5$ V/nm ($I \sim 0.3 \times 10^{12}$ W/cm^2 or 8 mJ/cm^2).

—: exp. ···: TDDFT



Resonant control on temporal emission dynamics of HHG



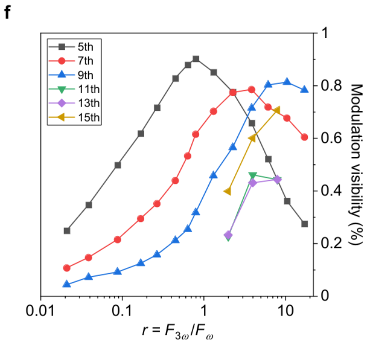
—: experiment | ···: TDDFT

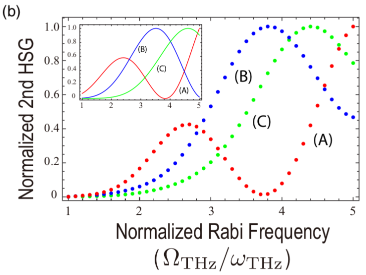
Our TDDFT predictions vs attosecond HHG experiment

Phase-dependent and time-dependent **agreement** for multiple harmonic orders in the **tunneling regime:** adequate to predict attosecond dynamics in solids.

HHG $\omega + 3\omega$ vs TDDFT: discussion

Gindl, A.; Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M.
Attosecond control of solid-state high harmonic generation using ω-3ω fields, arXiv:2310.07254
$\tau = 30$ fs, $\lambda_1 = 2000$ nm, $\lambda_2 = 666$ nm. $E \sim 1.5$ V/nm ($I \sim 0.3 \times 10^{12}$ W/cm^2 or 8 mJ/cm^2).





Tamaya, T. & Kato, T. *Phys. Rev. B*, **100**, 081203(R) (2019)

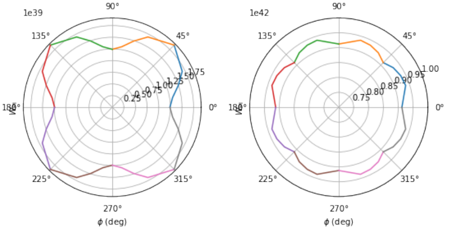
Discussion

Electron dynamics induced by bi-color mixing may be linked to the dynamical band structure (sub-bands & Rabi frequency variation with field & wavelength).

Outline

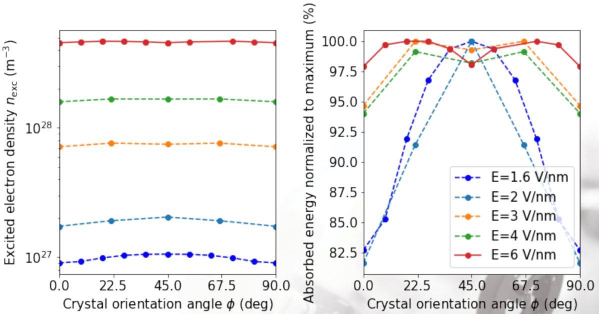
Outline

2023: TDDFT prediction of a ”new” control parameter: effect of Si [001] orientation



1 V/nm
(0.12 TW/cm²)

6 V/nm
(4 TW/cm²)



←: $n_{\mathrm{exc}}(\phi)$ excited e- density, →: $\xi_{\mathrm{abs}}(\phi)$ absorbed energy (eV)

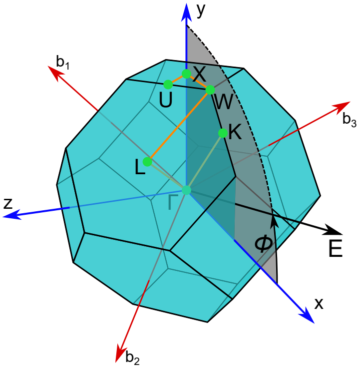
$E = 1.6$ V/nm ∼ 25 mJ/cm²
$E = 6$ V/nm ∼ 350 mJ/cm².

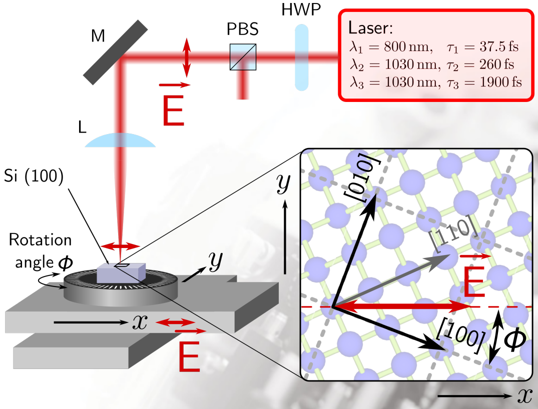
Orientation-dep. energy absorption pattern is also ... intensity-dependent

Low intensity: TDDFT predicts a 20% contrast in absorption pattern followinng the Si crystal symmetry.
High intensity: TDDFT predicts a 2% contrast (symmetry weakening / transition to plasma).

2023: Experimental protocol

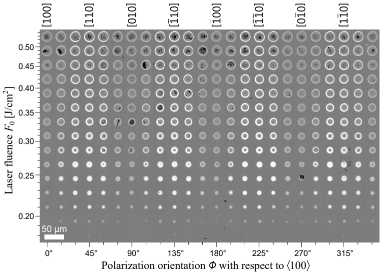
J. Sladek, **Y. Levy** (HiLASE Centre)

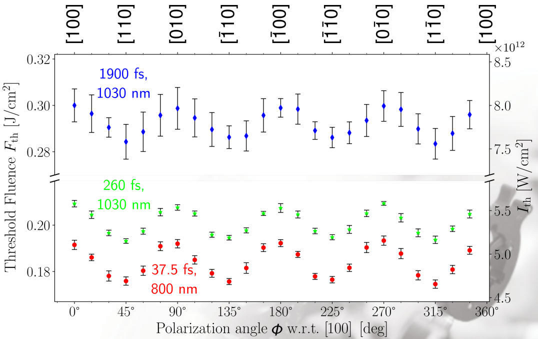




2023: Orientation-dep. amorphization threshold

Juraj Sladek & Yoann Levy (HiLASE Centre).





Condensed matter effect at various τ

Orientation-dependent damage threshold appears clearly, for 37 fs, 250 fs and 2 ps pulse duration.
[For <111>, see Florian et al., Materials **14**, 1651 (2021)].

Confirmed by Inst. de Optica (Madrid) & BAM Berlin

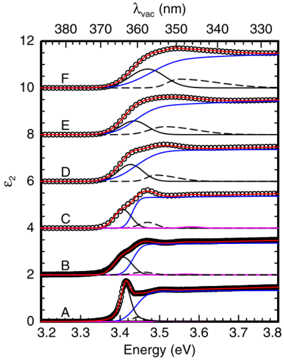
- **Invited stay** 15 days by Assoc. Prof. Mario Garcia-Lechuga (CSIC/iLINK project).

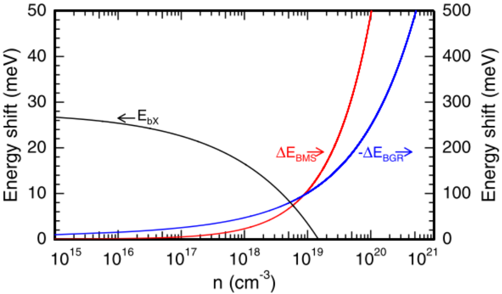
- **Experimental confirmation** of orientation-dependent damage threshold, pump-probe, ...

Press release (CSIC)

Discussion: Burnstein-Moss effect

Upon doping (resp. exciting electrons) a sample, band gap can shift (increase) due to saturation of band



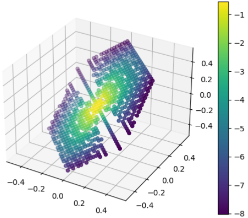


Feneberg, M. et al. Band gap renormalization and Burstein-Moss effect in silicon- and germanium-doped wurtzite GaN up to 10^{20} cm^{-3}, *Phys. Rev. B*, **90**, 075203 (2014)

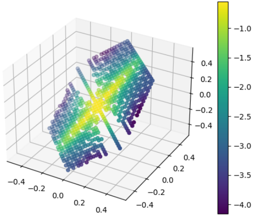
Burnstein-Moss effect?

Saturation phenomena should also depend on crystal orientation and laser polarization.

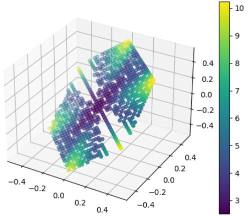
Discussion: Symmetry of Si bands $m_{\mathrm{eff}}(\phi)$ in [001]



Valence bands $\varepsilon(\mathrm{k})$ correspond. to plane [001] ("HOMO")



Valence bands $\varepsilon(\mathrm{k})$ correspond. to plane [001] ("HOMO")



Conduction bands $\varepsilon(\mathrm{k})$ correspond. to plane [001] ("LUMO")

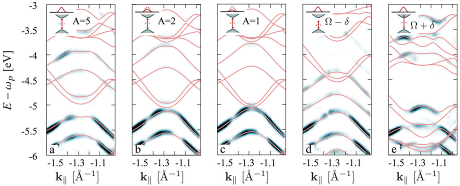
Outline

1. Context
2. TDDFT: multi-band description using high-power computations
3. How reliable are TDDFT predictions in the ultrafast regime? Benchmark vs high harmonic generation (HHG) experiments
4. Predictions of TDDFT at high intensity (laser processing)
5. Reversible and ultrafast band structure engineering
6. Overview: predictive modeling tools for high fields photonics are here and they work
7. IT4I/LQC: milestones towards designing a light-controlled multiqubit in solids?

Outline

Simulation of electron dressing in solids

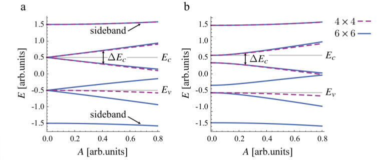
2016 De Giovannini, U.; Hübener, H. & Rubio, A. Monitoring Electron-Photon Dressing in WSe_2, *Nano Letters*, **16**, 7993-7998 (2016)



—: Floquet-Stark "dressed electron" energy levels.

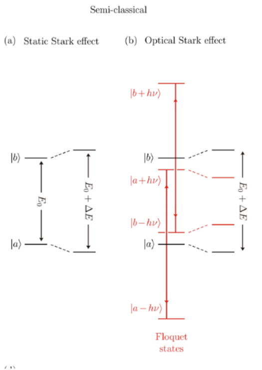
•: TDDFT-simulated angle-resolved photo-emission spectrum (ARPES)

Two-band Floquet (toy-)model as function of the laser field.



Theory for AC fields: "Floquet" $\mathscr{H}$

- Higuchi, T.; Stockman, M. I. & Hommelhoff, P. *Phys. Rev. Lett.* **113**, 213901 (2014)
- De Giovannini, U.; Hübener, H. & Rubio, A. *Nano Letters*, **16**, 7993-7998 (2016)
- Temporal integration of operators on 1 optical cycle: $\mathscr{H}(t) = \mathscr{H}(t + 2\pi/\omega)$.



$$H^{m,n}_{\text{Floquet}}(\omega) \equiv \int dt\, e^{-i(m-n)\hbar\omega t} H(t) \pm \delta_{m,n} n\hbar\omega$$

ω : laser photon energy (at. units),

n-photons transitions (number of "replicates"),

m: number of electronic bands,

$$H(t) = H_{\text{GS}} + \underbrace{\vec{A}(t)\cdot\bar{\bar{p}}}_{H_{\text{int}}}$$

A : vector potential amplitude (at. units)

$\bar{\bar{p}}$: momentum operator

$$H(t) = \underbrace{\begin{bmatrix} -E_g/2 & 0 \\ 0 & E_g/2 \end{bmatrix}}_{\text{ground state band structure at } k=(0,0,0)} + \underbrace{A\cos(\omega t)}_{\text{laser field}} \times \underbrace{\begin{bmatrix} 0 & M \\ \bar{M} & 0 \end{bmatrix}}_{\text{dipolar coupling matrix at } k=(0,0,0)}, \qquad (4)$$

M: dipolar transition matrix elements (at. units).

Example: $n = 1$ photon, $m = 2$ bands.

Coupling of dressed energy levels with **purely electronic levels**

$$H_{\text{eff}\,2\text{ bands}} = \begin{bmatrix} -\frac{E_g}{2}-\omega & -\omega & \cdot & \frac{AM}{2} & \cdot & \cdot \\ -\Omega & \frac{E_g}{2}-\omega & \frac{A\bar{M}}{2} & \cdot & \cdot & \cdot \\ \cdot & \frac{AM}{2} & -\frac{E_g}{2} & \cdot & \cdot & \frac{AM}{2} \\ \frac{A\bar{M}}{2} & \cdot & \cdot & \frac{E_g}{2} & \frac{A\bar{M}}{2} & \cdot \\ \cdot & \cdot & \cdot & \frac{AM}{2} & -\frac{E_g}{2}+\omega & \Omega \\ \cdot & \cdot & \frac{A\bar{M}}{2} & \cdot & \Omega & \frac{E_g}{2}+\omega \end{bmatrix}. \tag{5}$$

Eigen energy values (diagonalization):

$$E^{\text{eff}} = \begin{cases} \pm\frac{1}{2}\sqrt{2A^2 M\bar{M} + E_g^2} & \text{2 lev.} \\ \pm\frac{1}{2}\Big(\pm\sqrt{\bar{M}^2 A^4 M^2 + 16A^2 M\bar{M}\omega^2 + 64\omega^4 + 16\omega^2 E_g^2} + & \text{4 lev.} \\ +A^2 M\bar{M} + 8\omega^2 + E_g^2\Big) & \end{cases} \tag{6}$$

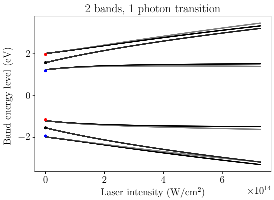
Conclusion for two-band models

In a two-band system, transient band-gap **increases** with field amplitude A (seems consistent with Keldysh "$\tilde{\Delta}$", "U_{eff}")

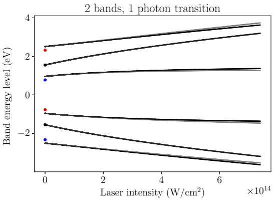
$$E_{\text{gap}}^{\text{eff}}(A) = \sqrt{2A^2 M\bar{M} + E_g^2} \propto A. \tag{7}$$

$m = 2$ bands, $n = 1$ photon.

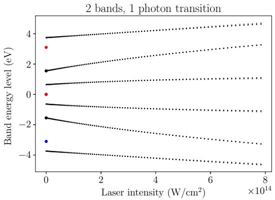
$E_g(\Gamma) = 2.56$ eV (LDA band gap of Si).



$\lambda = 3200$ nm



$\lambda = 1600$ nm



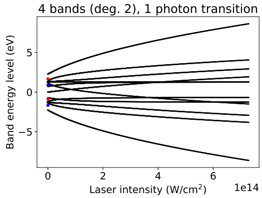
$\lambda = 800$ nm

Observation for 2 bands

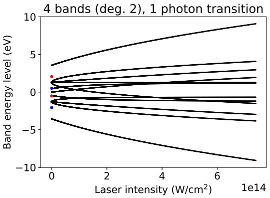
Shifting originates from the AC Stark effect. Opening of the band gap is observed.
Low intensity also evidences some ”detuning”.

$m = 4$ bands, $n = 1$ photon.

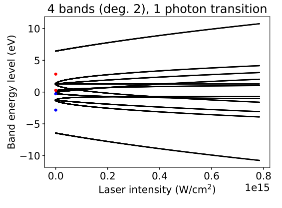
- In Si, 2 bands are degenerated 2 times at Γ-point. We use: $H(A=0) = \frac{1}{2}\begin{pmatrix} -E_g & \cdot & \cdot & \cdot \\ \cdot & -E_g & \cdot & \cdot \\ \cdot & \cdot & E_g & \cdot \\ \cdot & \cdot & \cdot & E_g \end{pmatrix}$



$\lambda = 3200$ nm



$\lambda = 1600$ nm



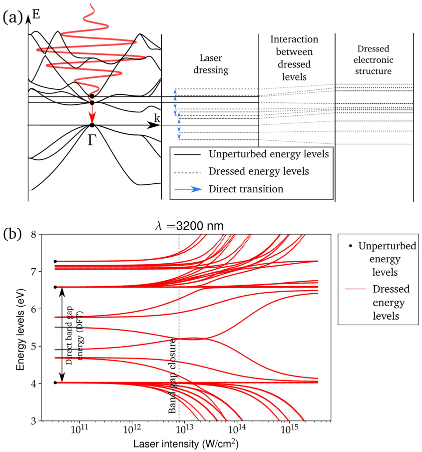
$\lambda = 800$ nm

Observation for 4 bands

Splitting of degenerated bands is observed. **Crossing** points are **observed** at points where band gap closes. This is laser-induced tunneling/metallization.

Application to Si: transient metallization at Γ

Effect of the laser excitation ($\vec{E}$ ↕) on the band structure?



T. J.-Y. Derrien, N. Tancogne-Dejean, [...] and N. M. Bulgakova, *Phys. Rev. B*. **104** L241201 (2021)

- Predictions for realistic materials is possible at **low cost** (10 min).
- Which materials undergo metallization / phase transition upon irradiation?

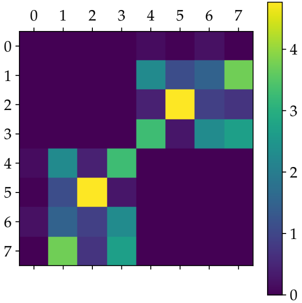
/!\ **Thermal effects are absent** in the presented descriptions.

Outline

Dipolar matrices

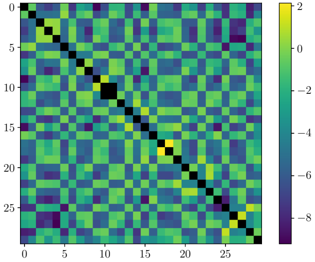
Subset of dipolar matrix for polarization in (Ox) direction.

Si [227], LDA (Γ: 2.56 eV)



Si, $k = 16^3$,
2 atoms, 8 electronic levels.

SiO_2 [154]: α-quartz



SiO_2, $k = 8^3$,
18 atoms, 30 electronic levels.

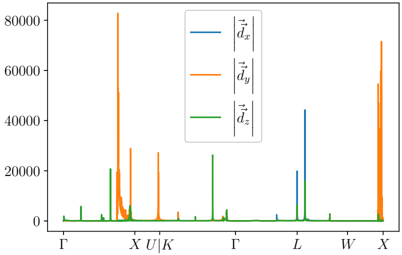
Density Functional Theory (DFT) - 3D

These results were computed for the 3D band structure.

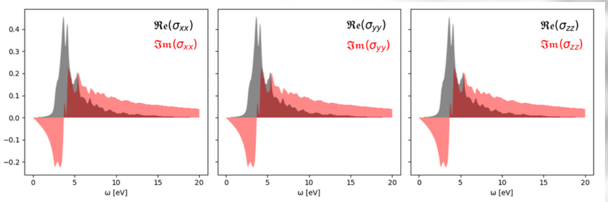
Validation of dipolar matrix elements

Lic. Andrés. I. Bertoni, Universidad de Cuyo (Mendoza, Argentina)

$|d_{i,j}|_{x,y,z}(k)$ for Si ($k = 24^3$, $\delta x = 0.18$, LDA).



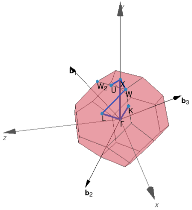
Linear response (AC conductivity) using Kubo-Greenwood (no Drude) as function of orientation. Built from $\vec{\vec{d}}_{x,y,z}$ for Si.

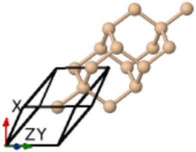


Outline

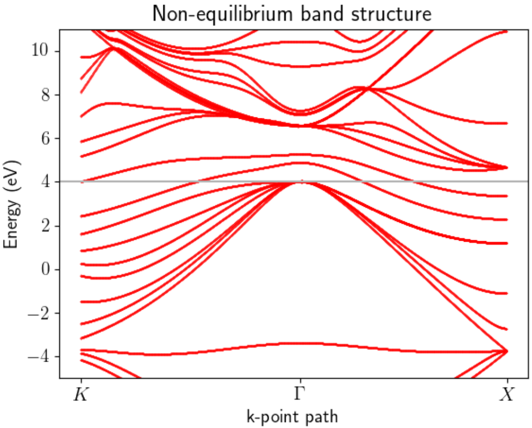
1. Context
   - Applications of ultrafast laser-induced phenomena in solids
   - Optical absorption driven by electrons dynamics in solids
     - *Frozen* band structure
     - *Dynamical* band structures: laser dressing was included in Keldysh (1965)
2. TDDFT: multi-band description using high-power computations
   - Modeling the laser excitation of electrons in Si (real-space, real-time TDDFT)
   - Scanning multiple parameters: database preparation
   - Results
3. How reliable are TDDFT predictions in the ultrafast regime? Benchmark vs high harmonic generation (HHG) experiments
4. Predictions of TDDFT at high intensity (laser processing)
   - TDDFT predictions for laser processing: anisotropy in energy absorption & damage threshold
5. **Reversible and ultrafast band structure engineering**
   - Simplified model: Floquet + DFT
   - Preparation of dipolar matrix elements (DFT)
   - **Si [227], LDA: $E_g^\Gamma = 2.56$ eV. Dressing along $K - \Gamma - X$**
   - Si [227], LDA: $E_g^\Gamma = 2.56$ eV. "3D" dressed band structure
   - Discussion
6. Overview: predictive modeling tools for high fields photonics are here and they work
7. IT4I/LQC: milestones towards designing a light-controlled multiqubit in solids?

Transient band structure: Si [227], E//(ΓX), $K - \Gamma - X$



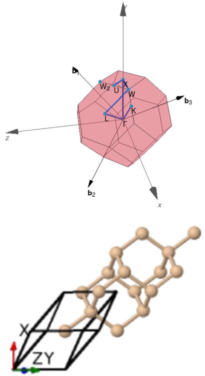


Orientation of
high-symmetry points
in real-space



$E = 0.001$ V/nm, $\lambda = 1030$ nm, $n = 1$

Transient band structure: Si [227], $E//(\Gamma X)$, $K - \Gamma - X$



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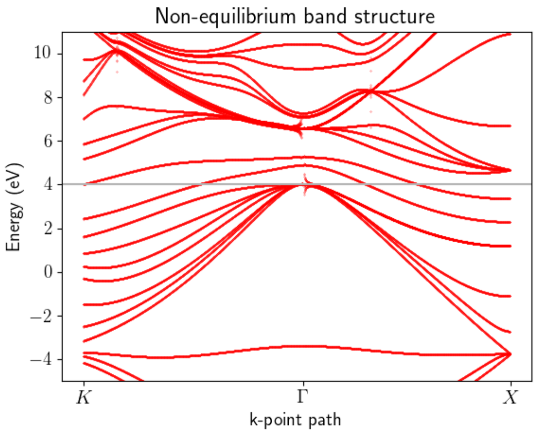

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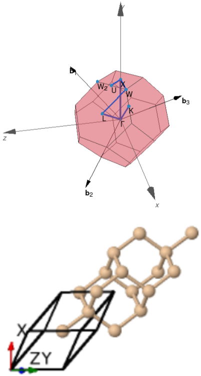
Transient band structure: Si [227], $E//(\Gamma X)$, $K-\Gamma-X$



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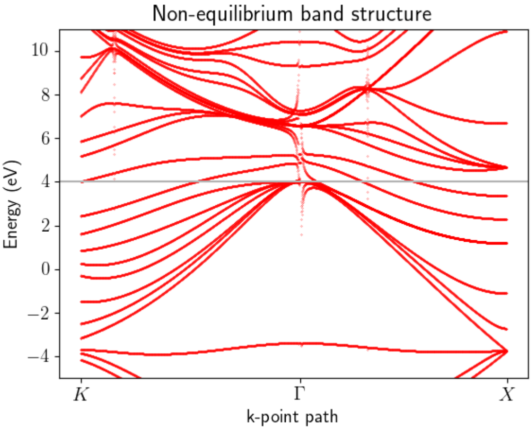

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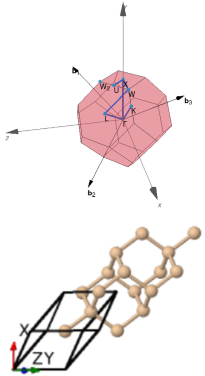

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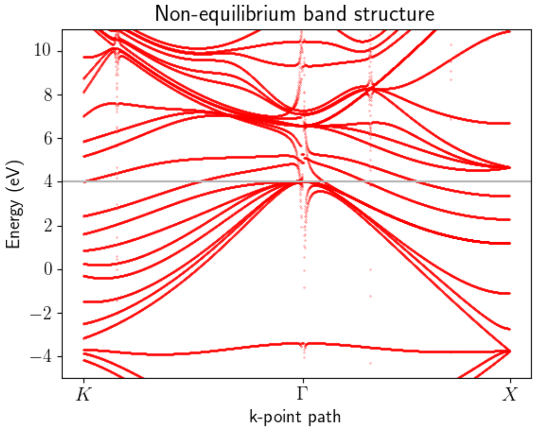

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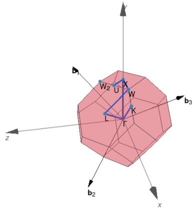
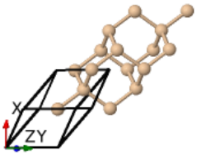
Transient band structure: Si [227], E//(ΓX), $K-\Gamma-X$





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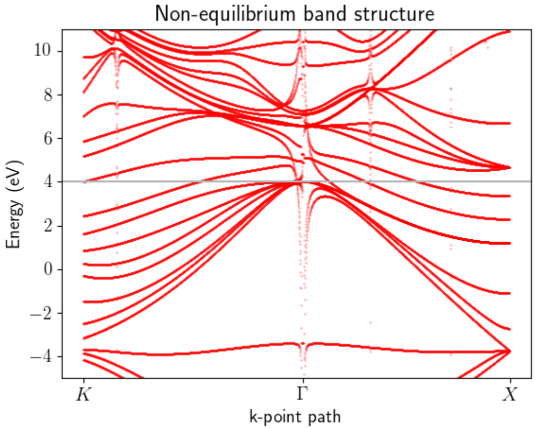

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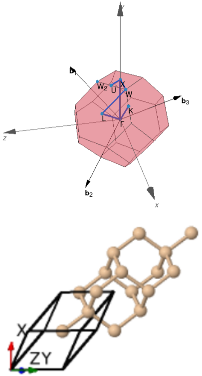
Transient band structure: Si [227], E//(ΓX), $K - \Gamma - X$



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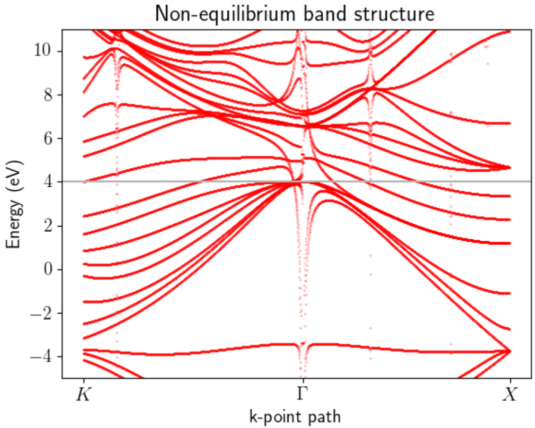

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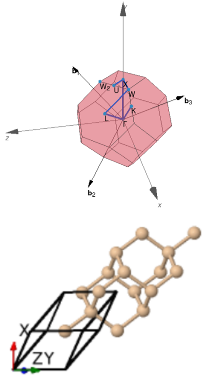
Transient band structure: Si [227], E//(ΓX), $K - \Gamma - X$



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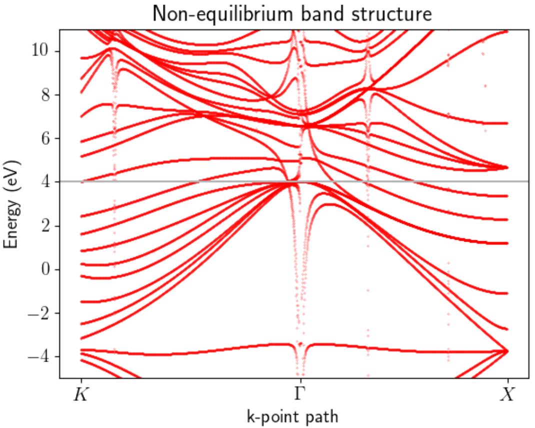

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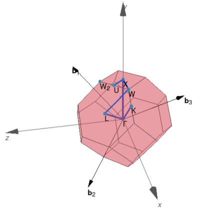
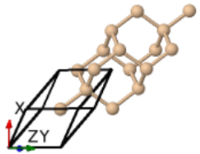
Transient band structure: Si [227], E//(ΓX), $K - \Gamma - X$





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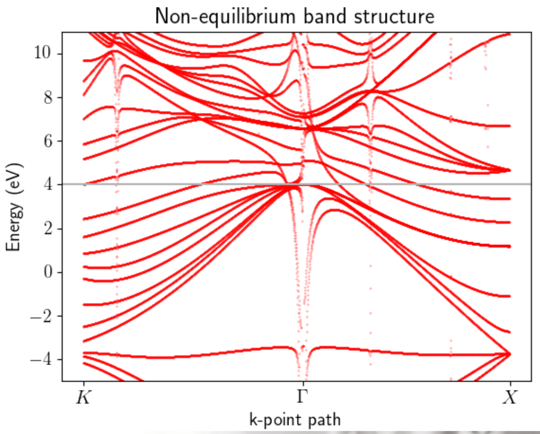
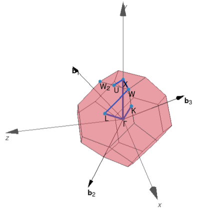
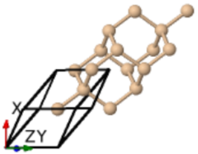


$E = 0.5$ V/nm, $\lambda = 1030$ nm, $n = 1$

Outline

Transient band structure: Si [227], $E//(\Gamma X)$





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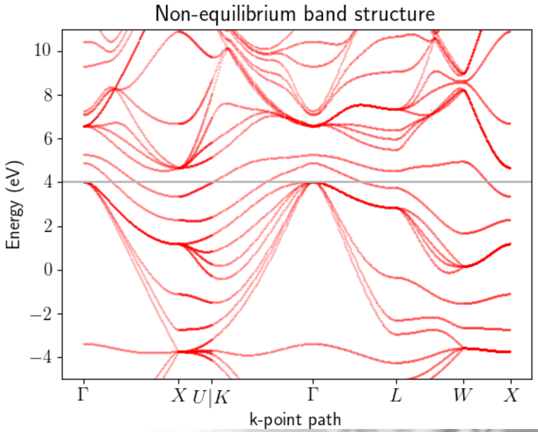

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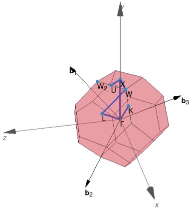
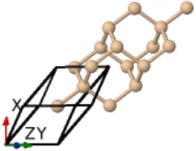
Transient band structure: Si [227], E//(ΓX)





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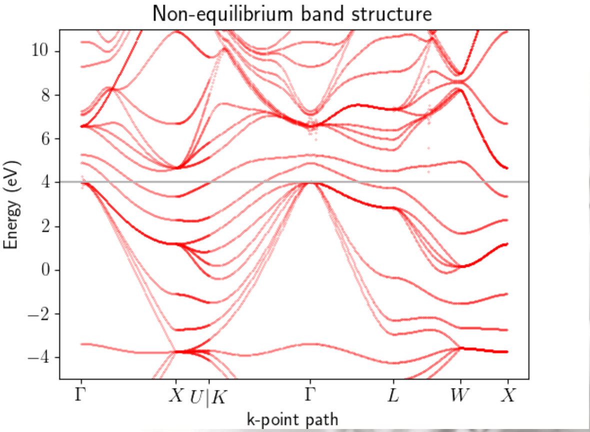

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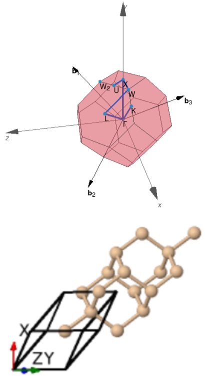

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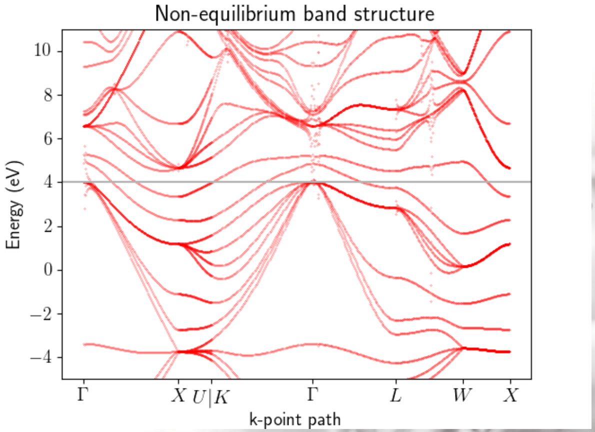

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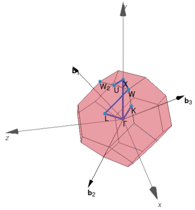

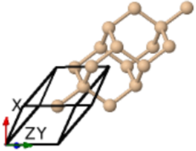

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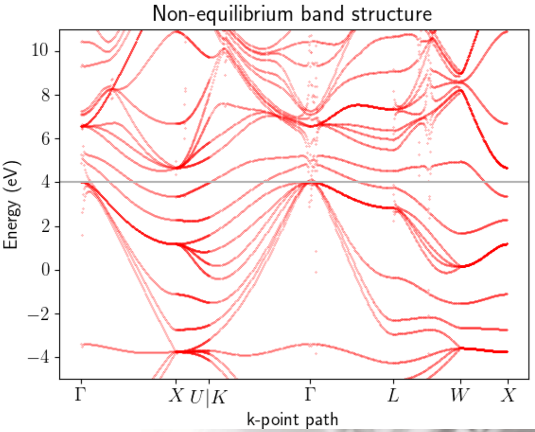

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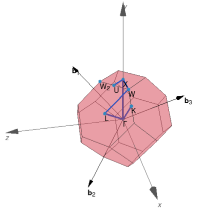

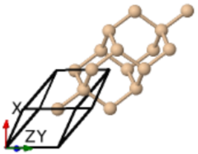

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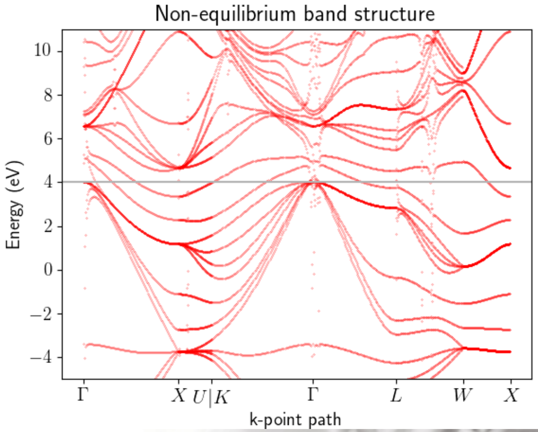

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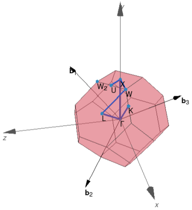

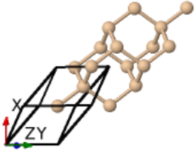

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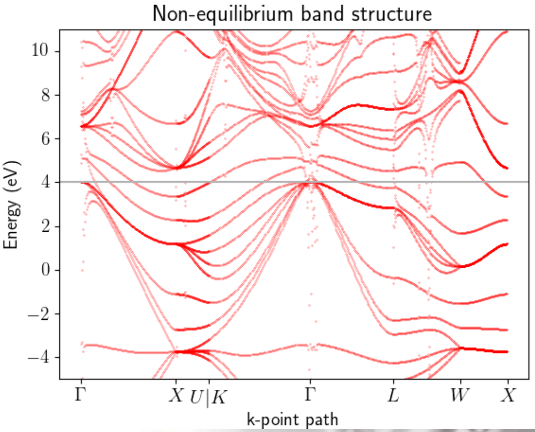

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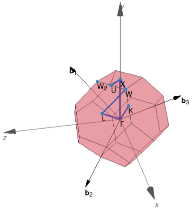

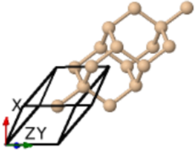

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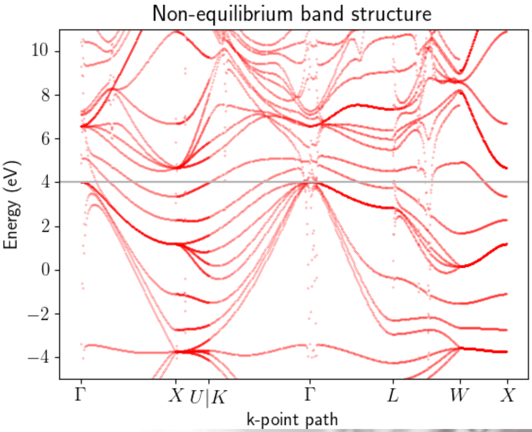

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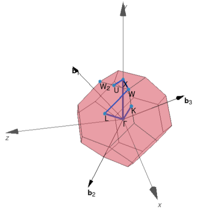

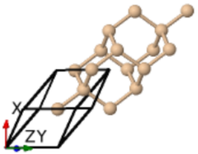

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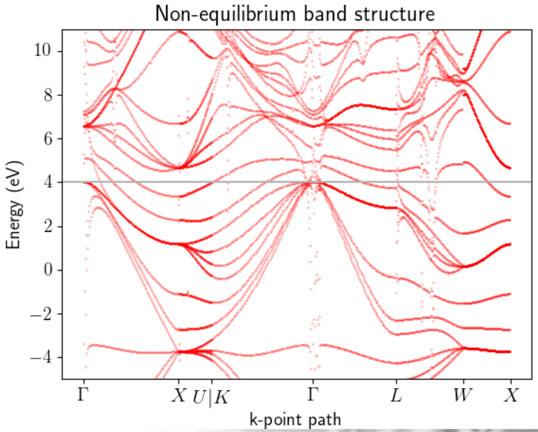
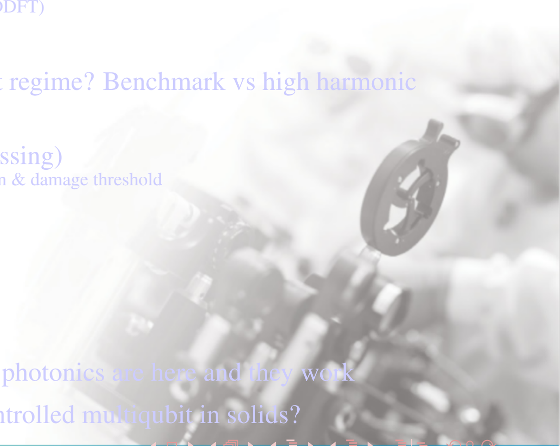


$E = 0.5$ V/nm, $\lambda = 1030$ nm, $n = 1$

Outline



Interpretation

Excitation direction	Response	No response
(ΓX)	(ΓX) $(K\Gamma)$ (ΓL) (WX) (LW)	(XU)

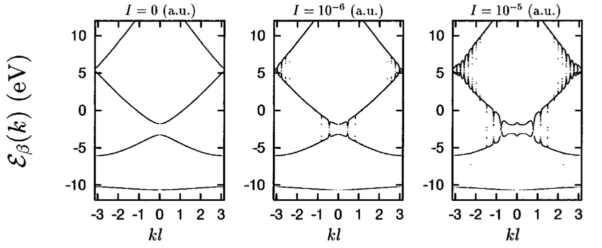
Interpretation

- ***Intraband* absorption** modifies band gap, and *modifies* interband absorption.
- **Strong *anisotropy*** *emerges from the laser irradiation*: constrained electron trajectories.

Discussion

Artefact? No!

Analytical models did exhibit a similar response in simplified cases.

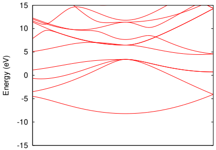


Faisal, F. H. M. & Kamiński, J. Z. Physical Review A **56**, 748-762 (1997)

Outline

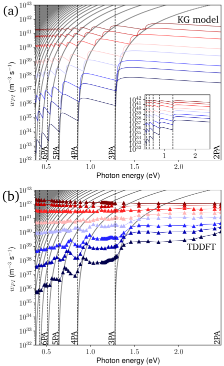
Predicting materials reaction to ultrafast light from quantum to large scales

Multiband quantum description of e- in matter



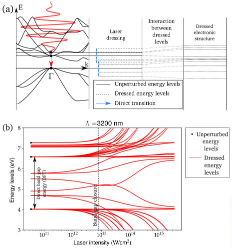
$K - \Gamma - X$

Benchmark of simplified theories



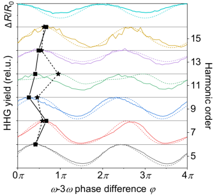
Derrien *et al.*, *Phys. Rev. B*. **104** L241201 (2021)

Transient band-gap dynamics



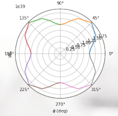
Derrien *et al.*, *Phys. Rev. B*. **104** L241201 (2021)

Benchmarking TDDFT vs reality (HHG)



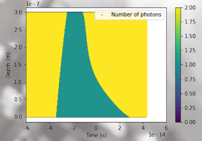
Suthar, P. et al. *Comm. Phys.* **5**, 288 (2022)
Gindl, A. et al. arXiv:2310.07254 (2024)

Predictions of new control parameters

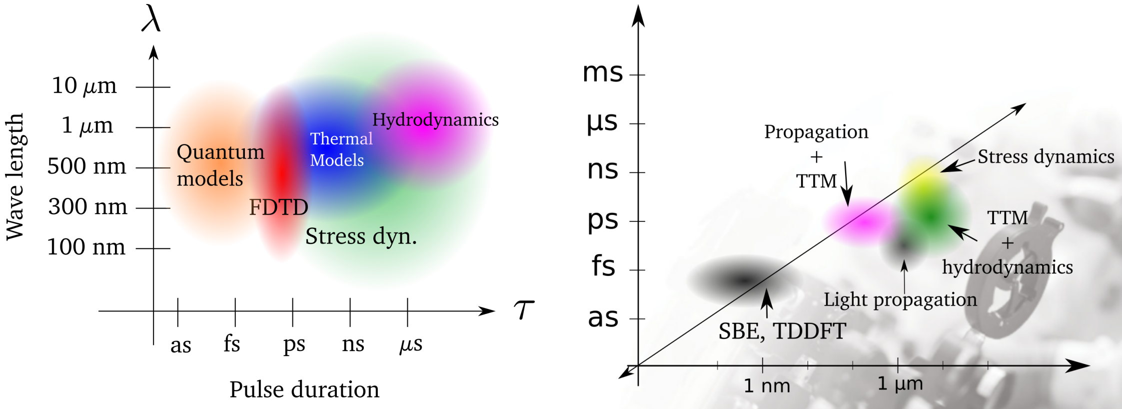


1 V/nm
(0.12 TW/cm²)

Quantum effects at large scales



Validity ranges of formalisms



Adadpted from proposal H2020-MSCA-RISE-2018 ”ATLANTIC”

Outline

1. Context
2. TDDFT: multi-band description using high-power computations
3. How reliable are TDDFT predictions in the ultrafast regime? Benchmark vs high harmonic generation (HHG) experiments
4. Predictions of TDDFT at high intensity (laser processing)
5. Reversible and ultrafast band structure engineering
6. Overview: predictive modeling tools for high fields photonics are here and they work
7. IT4I/LQC: milestones towards designing a light-controlled multiqubit in solids?

Milestones & outlooks

Considered done

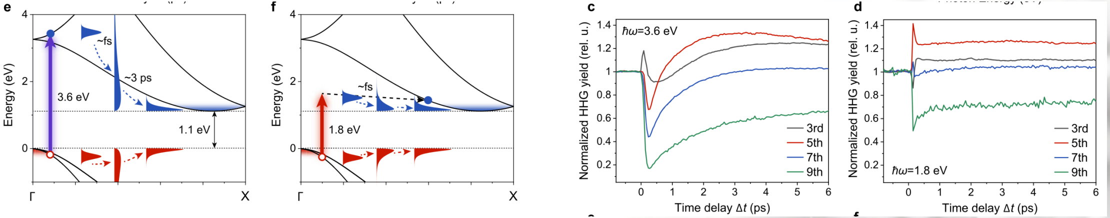
- Early instants (~ 50 fs) of light-matter interaction with **topologically trivial materials** (Si, SiO_2, SiC, CaF_2, ...) are now **well described** up to intensities enabling **materials modification** (strong field regime), still disregarding some induced defects: annealing, reduction of oxidation, etc...
- **Prediction capabilities** of available models at the ultrafast scales (faster-than-phonon response) have reached **excellent maturity**: demonstrated with HHG spectroscopy vs TDDFT.

The future: optimal control at ultrafast timescales

- **Ultrafast pulses** in perturbative regime should enable to use reversible population of exciton-polariton states (with fs/ps lifetimes), along with **avoiding damage**.
- **Occupations** of electronic states in matter can be populated at will (bicolor mixing).
- **Design crystals** and to consider **controlling their reaction upon few-cycle light**.

Relaxation dynamics: probing (in)direct transition using HHG(ω,t) spectroscopy

Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M. Momentum-dependent intraband high harmonic generation in a photodoped indirect semiconductor, *Comm. Phys.*, **7**, 104 (2024)



Need for relaxation dynamics

Finite temperature DFT + TDDFT possible.

What should be a good qubit?

Nielsen & Chuang 2010, p. 278, Quantum Computation & Quantum Information (Cambridge U. Press)

- √ **Finite number of states** (digital quantum computer): light intensity actually acts as a selector for the number of states.
- √ How to **probe the states**? Harmonic spectroscopy appears extremely precise, still state-destructive.
- √ **Timescales** of the interaction?
  - τ_Q : time of decoherence
  - τ_{op} : time for population of states
  - fs laser + Si = FAST! but not a great coherence time.

⟶ Multi-material study @ poster "Quantum Dynamics of Systems" @ IT4I Users Meeting, November.

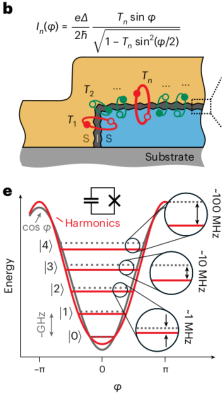
System	τ_Q	τ_{op}	$n_{op} = \lambda^{-1}$
Nuclear spin	$10^{-2} - 10^{8}$	$10^{-3} - 10^{-6}$	$10^{5} - 10^{14}$
Electron spin	10^{-3}	10^{-7}	10^{4}
Ion trap (In^{+})	10^{-1}	10^{-14}	10^{13}
Electron – Au	10^{-8}	10^{-14}	10^{6}
Electron – GaAs	10^{-10}	10^{-13}	10^{3}
Quantum dot	10^{-6}	10^{-9}	10^{3}
Optical cavity	10^{-5}	10^{-14}	10^{9}
Microwave cavity	10^{0}	10^{-4}	10^{4}

Exciton-polaritons in Si as multiqubits? Not great, but fast!

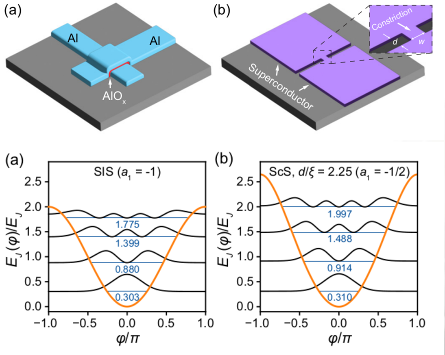
$\tau_Q \sim \tau_{\text{e-ph}} \geq 5 \times 10^{-14}$ s | $\tau_{op} \sim 5 \times 10^{-17}$ s | $n_{op} \sim 10^{3}$.

Superconducting transmon & geometry

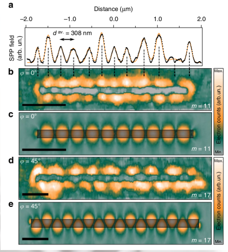
Willsch et al. Nat. Phys. **20**, 815 (2024)



Liu and Black, Phys. Rev. A **110**, 012427 (2024)



Piazza et al., *Nature Communications* **6**, 6407 (2015)



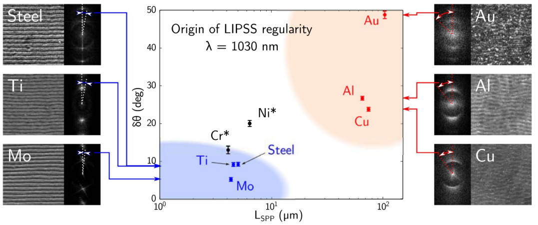
Next steps: classical multi-material approach

July 2016 Patent in CZ.

Aug. 2017 Publication in *Scientific Reports* (Nat. Publ. Group).

Nov. 2017 Press articles: Technical Weekly, Ceska Televize (CTV 24), Novinky.cz.

Jan 2018 Patent in EU No. WO2018010707.

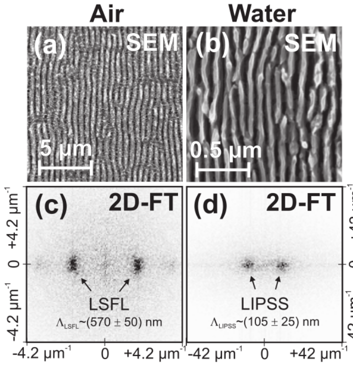


Gnilitskyi, I.; Derrien, T. J.-Y., Levy, Y., Bulgakova N. M. et *al. Sci. Rep.* **7**, 8485 (2017).

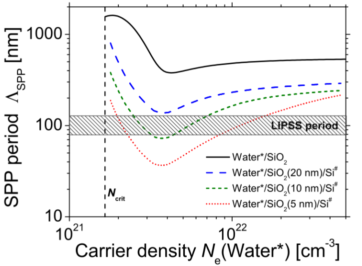
What was key?

A multi-material approach enabled by databasing of optical properties, and of plasmon polaritons properties.

Polaritons in thin films

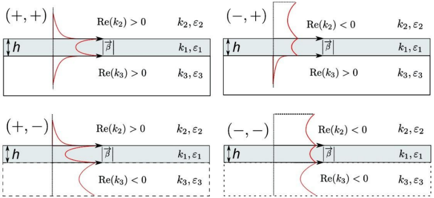


Derrien, T. J.-Y.; Koter, R.; Krüger, J.; Höhm, S.; Rosenfeld, A. & Bonse, J.; *J. Appl. Phys.*, **116**, 074902 (2014)



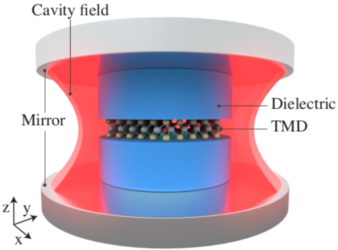
Derrien, T. J.-Y.; Koter, R.; Krüger, J.; Höhm, S.; Rosenfeld, A. & Bonse, J.; *J. Appl. Phys.*, **116**, 074902 (2014)

Strong coupling polaritonics



A. Dostovalov, **T. J.-Y. Derrien,** S. Lisunov, [...] and N. M. Bulgakova, *Appl. Surf. Sci.* **491**, 650 (2019).

Latini, S.; Ronca, E.; de Giovannini, U.; Hübener, H. & Rubio, A., Nano Letters **19**, 3473 (2019).



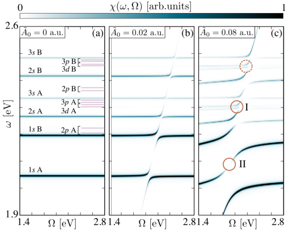
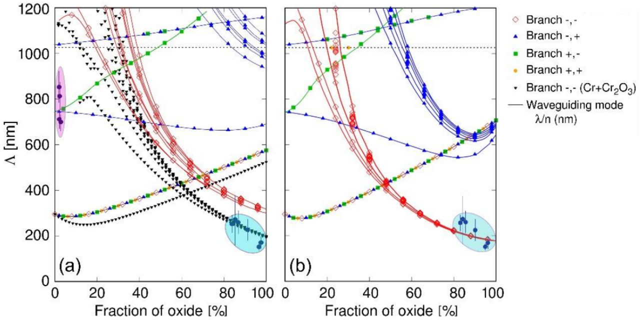


FIG. 2. Exciton-polariton spectra of MoS_2 in a cavity as a

Multiple polaritons in thin cavities



A. Dostovalov, **T. J.-Y. Derrien,** S. Lisunov, [...] and N. M. Bulgakova, *Appl. Surf. Sci.* **491**, 650 (2019).

High-spatial frequency LIPSS explained by SPP, oxidation and porosity.

Multiple polaritonic modes are found in metal and in oxides.
LSFL-|| are well explained by Plasmon Polariton in metallic regime.
Oxidation seems to play a major role in the formation of HSFL-⊥, $\Lambda \ll \lambda$ structures.

IQM (Finland) won the tender!

Press release September 26th, 2024

VSB TECHNICAL UNIVERSITY OF OSTRAVA | IT4INNOVATIONS NATIONAL SUPERCOMPUTING CENTER

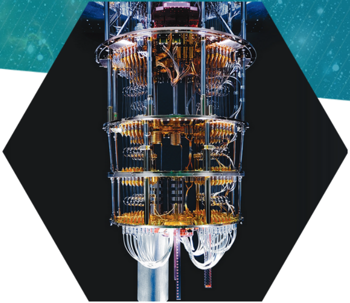
EUROCC E-INFRA CZ EXTRANET | CZ

ABOUT INFRASTRUCTURE RESEARCH INDUSTRY COOPERATION FOR USERS EDUCATION EVENTS

ABOUT → INFOSERVICE → NEWS → EUROPE TAKES A QUANTUM LEAP: LUMI-Q CONSORTIUM SIGNS CONTRACT TO ESTABLISH QUANTUM COMPUTER IN THE CZECH REPUBLIC

EUROPE TAKES A QUANTUM LEAP: LUMI-Q CONSORTIUM SIGNS CONTRACT TO ESTABLISH QUANTUM COMPUTER IN THE CZECH REPUBLIC

Luxembourg, 26 September 2024 – The European Union has taken another step towards enhancing the European quantum computing infrastructure by signing a contract for the acquisition of the LUMI-Q consortium's quantum computer. The quantum computer will be housed in Ostrava, Czech Republic, at the IT4Innovations National Supercomputing Center, part of the VSB – Technical University of Ostrava. The contract was signed between the European High-Performance Computing Joint Undertaking (EuroHPC JU) and IQM Quantum Computers, the company selected to supply the unique technology.



Collaborations & publications

thibault.derrien@vsb.cz | derrien@fzu.cz | Twitter/X @tjyderrien

Current financial support

- FZU Institute of Physics, Prof. N. M. Bulgakova. Sendiso project.
- IT4Innovation, V. Vondrak, B. Jansik, M. Lampart.

Experimentalists

- assoc. prof. Martin Kozák, Charles University (Prague, Czech Republic).
- Dr. Yoann Levy & Dr. Juraj Sladek, HiLASE Centre (Prague, Czech Republic).
- Dr. Jörn Bonse, BAM (Berlin, Germany).

Theoretical support

- HiLASE Centre: PhD std. Kristyna Gazdova
- UNCuyo, Mendoza, Argentina : Lic. Andrés I. Bertoni, Prof. Cristián Sanchez.
- MPSD Hamburg: Dr. Nicolas Tancogne-Dejean, Dr. Franco Bonafé

Selected (quantum) publications of this presentation

TDDFT Si Derrien, T. J.-Y.; Tancogne-Dejean, N.; Zhukov, V.; Appel, H.; Rubio, A. & Bulgakova, N. M. *Phys. Rev. B* **104**, L241201 (2021).

More details on TDDFT Derrien, T. J.-Y.; Levy, Y. & Bulgakova, N. M. Chap. 1 in *Ultrafast Laser Nanostructuring - The Pursuit of Extreme Scales* (Eds. R. Stoian, J. Bonse), Springer, 2023.

HHG Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M., *Comm. Phys.* **5**, 288 (2022).
Gindl, A.; Suthar, P.; Trojánek, F.; Malý, P.; Derrien, T. J.-Y. & Kozák, M., arxiv:2310.07254.

Orientation Sládek, J.; Levy, Y.; Bonse, J.; Bulgakova, N. M. & Derrien, T. J.-Y. Polarization-dependent damage threshold of Si [100] upon femtosecond and picosecond laser irradiation. *In finalization*.

QuantumLaP.eu

GROUP OF ULTRAFAST PHOTONICS (T. J.-Y. DERRIEN)

LATEST NEWS | GROUP MEMBERS | REVIEWS | PREDICTION TOOLS | COMMUNICATIONS | FUNDING

HIGH POWER COMPUTATION | CONTACT US

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Schleder G. R., Padilha A. C. M., Acosta C. M., Costa M., Fazzio A., 2019, Journal of Physics: Materials, 2, 032001

Stefanucci G., Leeuwen R. v., 2013, Nonequilibrium many-body theory of quantum systems: a modern introduction. Cambridge University Press, Cambridge

Tancogne-Dejean N., Mücke O. D., Kärtner F. X., Rubio A., 2017, Nature Communications, 8, 745

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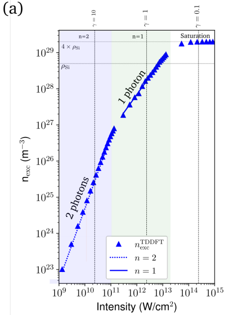
Willsch D., et al., 2024, Nat. Phys., 20, 815

Outline

8. Re-using TDDFT datasets: introduce the TDDFT excitation rates into large scale description (rate equations)

9. Supplementary slides

Excitation of electrons: band gap reduction & data continuity



Derrien, T. J.-Y.; Tancogne-Dejean, N.; Zhukov, V.; Appel, H.; Rubio, A. & Bulgakova, N. M. *Phys. Rev. B*, **104** L241201 (2021)

A data approach to electron excitation

- Multiphotonic rates are not directly usable: number of photons n changes with intensity (ultrafast metallization)

$$\sigma_n I^n \rightarrow \sigma_{n(I)} I^{n(I)}.$$

- Keldysh model is not directly usable: it has discontinuities due to interruption of transition between 2 levels (”Wannier-Stark localization”).
- Data from TDDFT are contiguous, due to multiband description: only 1 transition can be disabled at once (theorem of ”Le Bourget”).

Key point

In atomic physics, n *increases* with intensity.
In solid state physics, n *decreases*.

Let’s try!

Reduction of the band-gap during pulse → **effect at large spatial scale** (μm)?

1st attempt: sources terms for rate equations (RE)

- Density of conduction bands electrons

$$\frac{\partial n_{\text{exc}}}{\partial t} + \nabla \cdot \mathrm{J} = G_{\text{e-h}} + R_{\text{AR}} \tag{8}$$

$$G_{\text{e-h}} = \left\{ w_{\text{PI}}^{\text{TDDFT}} \left[f\left(\text{Re}\sqrt{\varepsilon_{\text{eq}}(\omega)}\right) \times I(t) \times [1 - R(t)] \right] + \right.$$
$$\left. + \underbrace{\delta_{\text{II}}(n_{\text{exc}})}_{\text{negligible if } \tau \ll \tau_{\text{eph}}} \times n_{\text{exc}}(t) \right\} \times \frac{n_0^{\dagger} - n_{\text{exc}}(t)}{n_0^{\dagger}} \tag{9}$$

where $f(x) = x \times \theta(x)$ and $\theta(x)$ is Heaviside function.

- Beer-Lambert law

$$\frac{dI}{dz} = -\left(\hbar\omega \times n_{\text{ph}}^{\text{TDDFT}}(I) \times \underbrace{w_{\text{PI}}^{\text{TDDFT}}[I]}_{\text{MPI+tunnel+fcr. abs.}}\right), \tag{10}$$

Method for interfacing

Difficulties

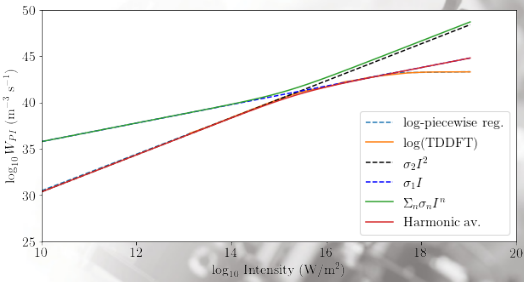
- Discontinuity of the Keldysh model at Wannier-Stark localizations
- Number of photons for multiphoton absorption: depends on intensity!

$$n = n(I)$$

- TDDFT provides predictions for high intensity pulse: how to describe intensities from 0 to our calculations?

Methods

- Extend the TDDFT data down to 0 V/m by using piece-wise interpolation.
- Directly use the resulting parametrization inside the RE model, and address various wavelengths.



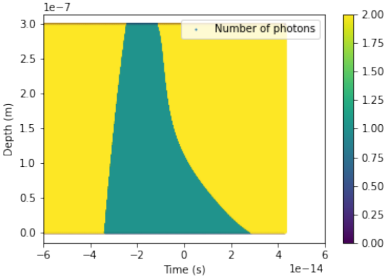
Large-scale effects of dressing (λ =800 nm, τ =30 fs)

Spatio-temporal evolution of number of photons necessary for a direct transition in Si.

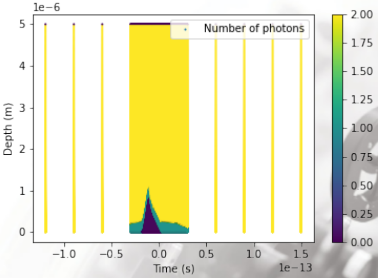
Introducing the laser dressing into large-scale description

Laser modifies the gap of interaction *during* the pulse.

0.1 µm thin Si sample - 0.18 J/cm²



5 µm thick Si sample - 0.5 J/cm²

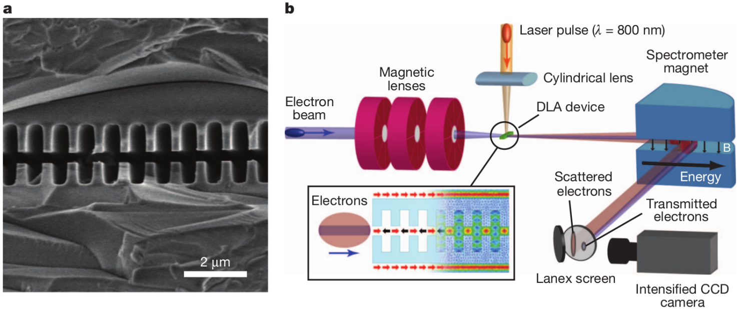


Band structure engineering using light

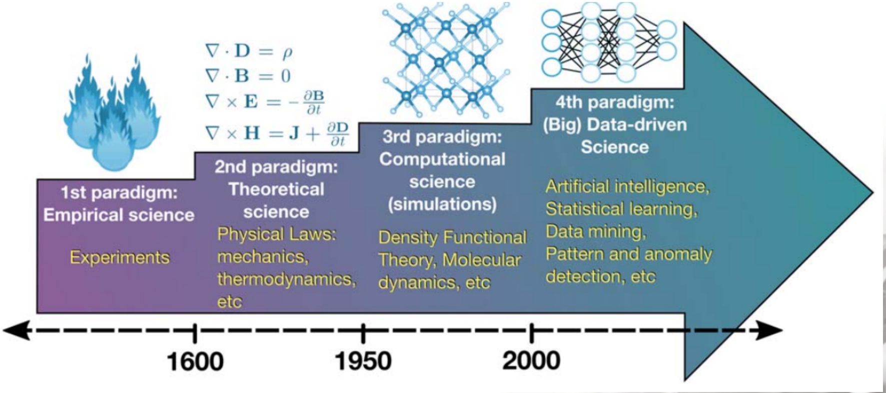
Large-scale consequence of laser dressing: a position-dependent band structure ⇒ ultrafast currents generated in the band gap material → electron acceleration.

Miniaturisation of accelerators using dielectrics gratings and fs lasers

Peralta, E. A.; Byer, R. L. et al, *Demonstration of electron acceleration in a laser-driven dielectric microstructure,* Nature **503**, 91 (2013)



Research frameworks



(Schleder et al., 2019)

Quantum approaches in the production cycle

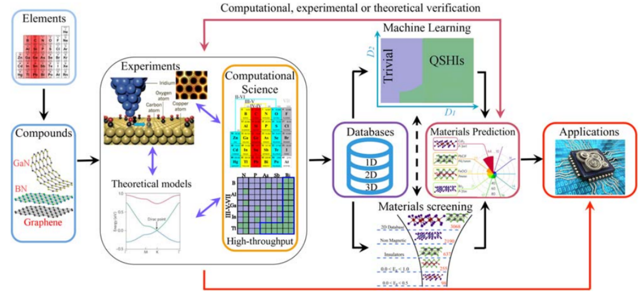
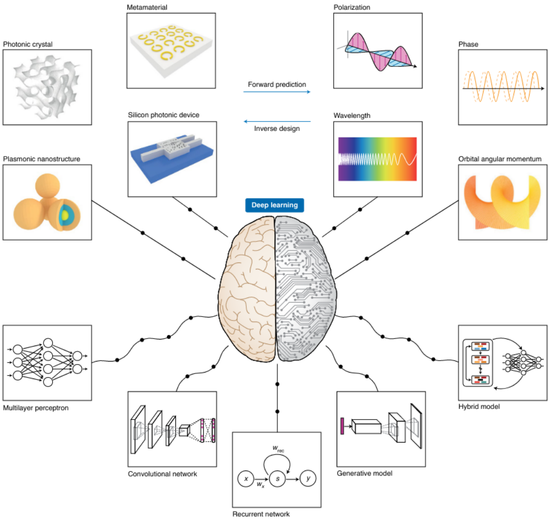


Figure 1. Schematic presentation of the topics discussed in this review and their relationships. The possible atomic combinations form a great number of compounds, which can be studied by means of experimental, theoretical or computational approaches, especially with high-throughput calculations. Large quantities of data generated are then stored in databases, which can be used by means of materials screening or machine learning, both of which leads to promising materials candidates. Data-driven or traditional routes select materials suited for specific applications. We illustrate these relationships in the context of two-dimensional materials. Figures from panels adapted with permission from [1] (experiments) (© 2012 Macmillan Publishers Limited. All rights reserved. With permission of Springer), [2] (computational science and machine learning), and [3] (materials prediction) (© 2018 Macmillan Publishers Limited, part of Springer Nature. All rights reserved. With permission of Springer).

Schleder, Gabriel "From DFT to machine learning: recent approaches to materials science", *Journal of Physics: Materials* **2**, 032001 (2019).

Pulse shape optimization + photonic design



Ma, Boltasseva et al (Ma et al., 2021)

Special session @ LPM 2023, Japan

Special session "Machine learning and Simulation for Laser Processing"

- Home & news
- Topics
- Special sessions
- Invited speakers
- Technical digest
- Advance program

Welcome to LPM2023

The 24th International Symposium on Laser Precision Microfabrication
Hirosaki Bunka Center, Hirosaki, Aomori, Japan
June 13-16, 2023

LPM2023 -The 24th International Symposium on Laser Precision Microfabrication will be held from June 13 to June 16, 2023 as "**In-Person**" conference in Hirosaki-city, Aomori prefecture, Japan.

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Special Session 2: Machine Learning and Simulation for Laser Processing

Session organizers:

Prof. Kenichi Ishikawa, The University of Tokyo, Japan

Dr. Tomohito Otobe, National Institutes for Quantum and Radiological Science and Technology, Japan

Dr. Thibault J.-Y. Derrien, HiLASE Centre, Czech Republic

Short description:

Laser processing is flexible with many tunable parameters such as wavelength, pulse duration, pulse energy, and scan speed. Today, these parameters are optimized by human experience and intuition. In this special session, to meet the mass customization need in the coming super smart society, we discuss alternative approaches driven by data, artificial intelligence, and numerical simulations

Database of excitation rates, absorbed energy, ...

For Si, ~3,000 TDDFT simulations with relevant laser pulses have been prepared [~2.7 M-core-hours per year]

- Several materials (Si, Mo, Au, ...)
- Several pulse shapes, pulse mixtures, ...
- Several observables (absorbed energy, currents, harmonic spectra, ...).
- All the work has been systematized into PYTHON & BASH routines for collaboration purposes.

High Power Computation Projects

- **IT4Innovations** National Supercomputing Center - eINFRA (ID:90140), sub-proj. MORILLE, FLAMENCO, FILIPINAS.
- **PRACE** aisbl (projects BOLERO, FRECUENCIA).

Backup National Grid Infrastructure **MetaCentrum** eINFRA (ID:90140).

VŠB TECHNICKÁ UNIVERZITA OSTRAVA | IT4INNOVATIONS NÁRODNÍ SUPERPOČÍTAČOVÉ CENTRUM

ML with quantum approaches

Open questions

- What could be done beyond interpolation / extrapolation of existing results?
- Can it help to reduce k-grid space? mesh space? decrease cost of calculations?
- How many TDDFT simulations are necessary to train the algorithm? millions? thousands?

Attempts using SKLEARN for now: extrapolation of n_{exc} from $\sim$18 input parameters.

Talent competition high-school student (CZ)

Topic: Supervised machine-learning on existing TDDFT datasets for accelerating laser processing.

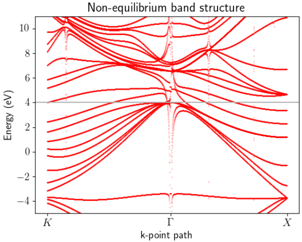
Preliminary results

A satisfactory training requires ~100^+ TDDFT data points in a given set of parameters.

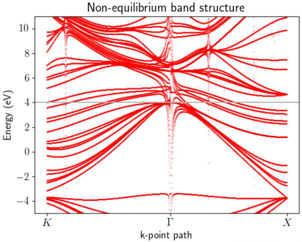
Outline

Contributions of higher orders

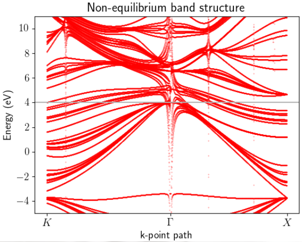
$n = 1, 2, 3, \ldots$ replicates. $E = 0.2$ V/nm, $\lambda = 1030$ nm.



$n = 1$



$n = 2$



$n = 3$