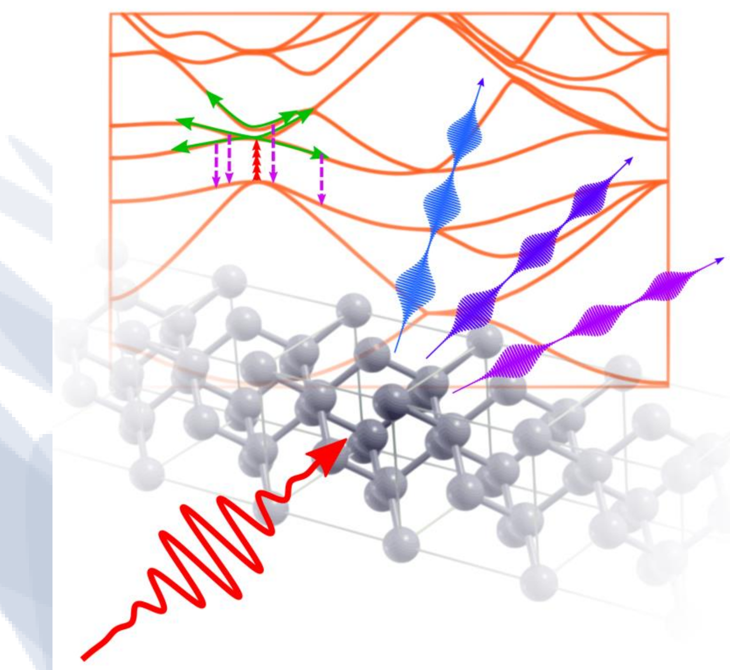
High-Harmonic Generation from solids and two-dimensional materials

Nicolas Tancogne-Dejean

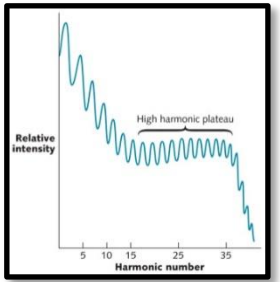


Collaborators: O. D. Mücke, F. X. Kärtner, Angel Rubio

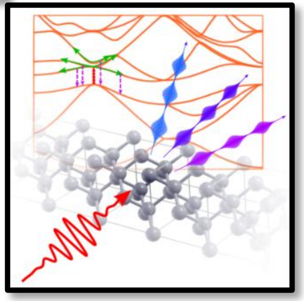
Max Planck Institute for the Structure and Dynamics of Matter

Outline

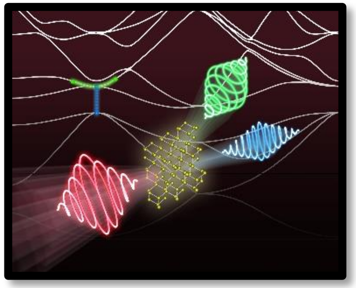
- High-harmonic generation (HHG)



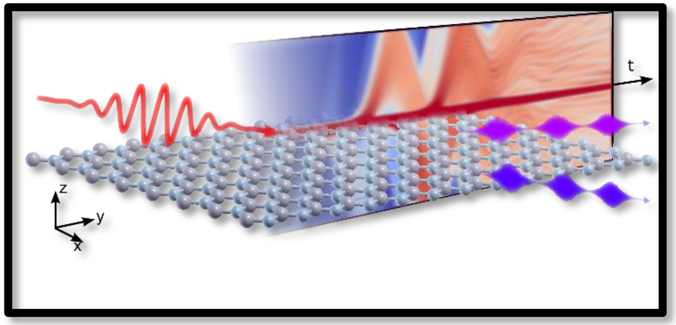
- Impact of the band-structure



- Ellipticity dependence



- Atomic-like HHG from 2D materials



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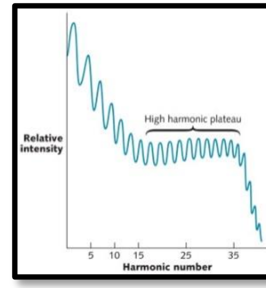

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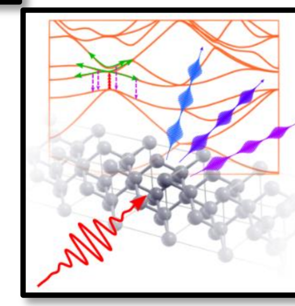

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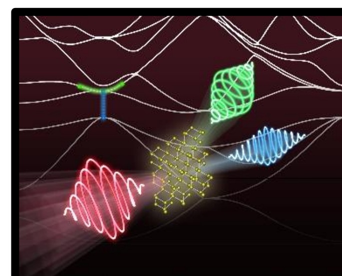

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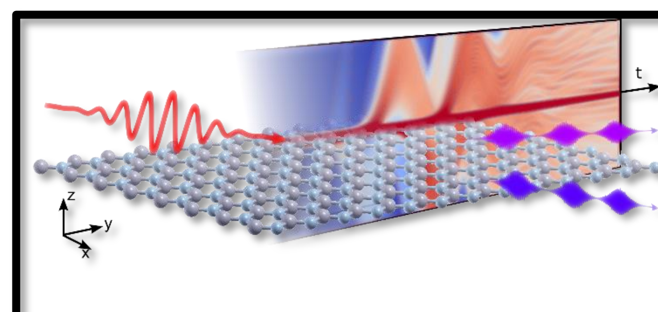


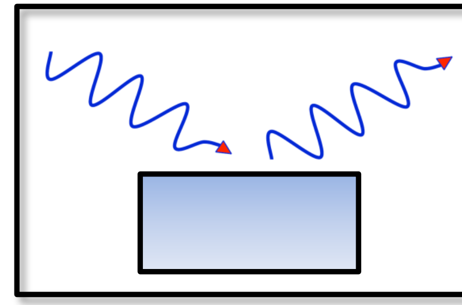
Response to a perturbation

Perturbation

Electric field

Response

Polarisation



Linear Response | **Nonlinear Response**

$$P_i = \boxed{\sum_j \chi^{(1)}_{ij} E_j} + \boxed{\sum_{jk} \chi^{(2)}_{ijk} E_j E_k + \sum_{jlk} \chi^{(3)}_{ijkl} E_j E_k E_l + \ldots}$$

Response to a perturbation

mpsd

Perturbation

Electric field

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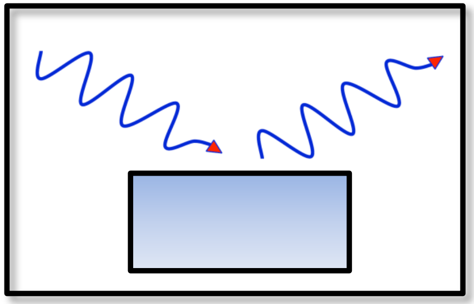
Polarisation



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For weak lasers
($< 10^{11}$ W/cm^2)

$$\chi^{(1)} E \gg \chi^{(2)} EE \gg \chi^{(3)} EEE \gg \ldots$$

Perturbative regime

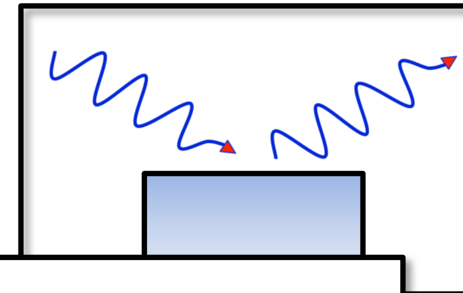
Response to a perturbation

Perturbation

Electric field

Response

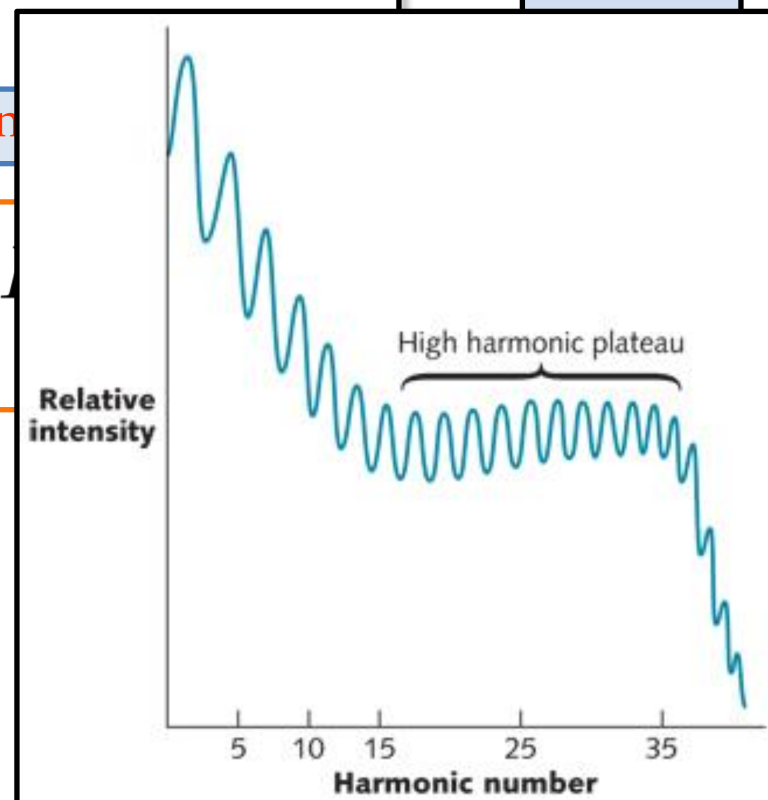
Polarisation



Linear Response

Non

$$P_i = \boxed{\sum_j \chi^{(1)}_{ij} E_j} + \boxed{\sum_{jk} \chi^{(2)}_{ijk} E_j \dots} \dots$$



For strong lasers

($> 10^{11}$ W/cm^2)

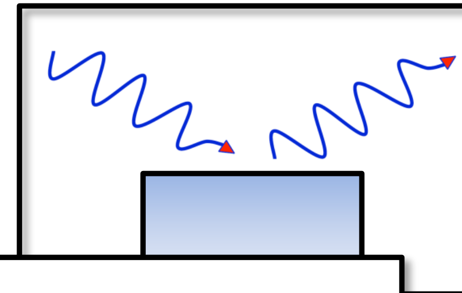
Response to a perturbation

Perturbation

Electric field

Response

Polarisation



Linear Response

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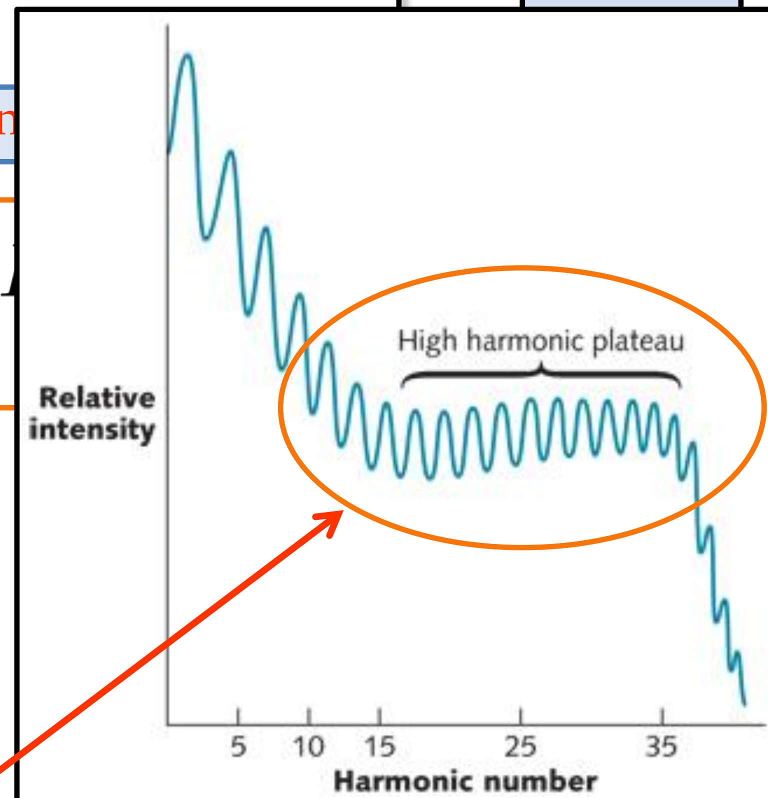
$$P_i = \boxed{\sum_j \chi^{(1)}_{ij} E_j} + \boxed{\sum_{jk} \chi^{(2)}_{ijk} E_j} \dots$$

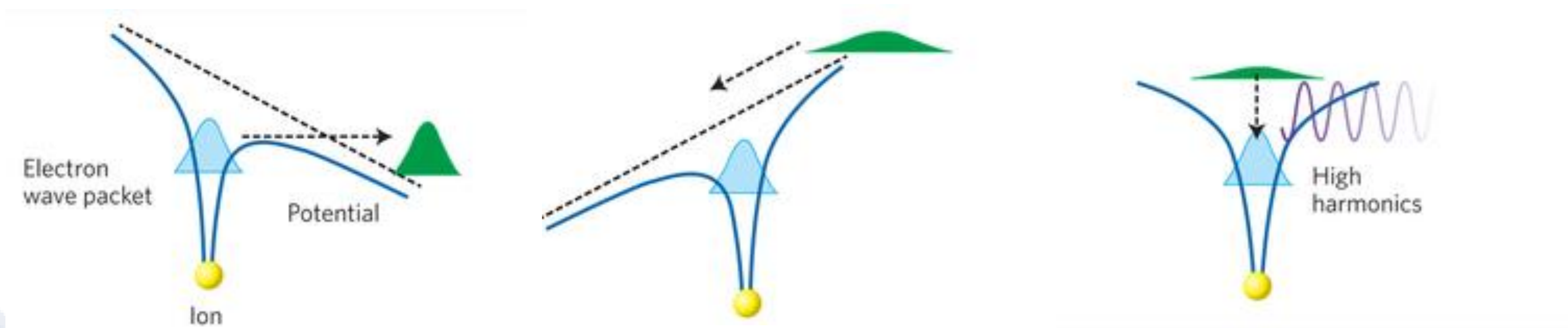


For strong lasers
($> 10^{11}$ W/cm^2)

Non-perturbative regime

HHG in atoms: three-step model

HHG in atoms is well explained by the three-step model [1,2]

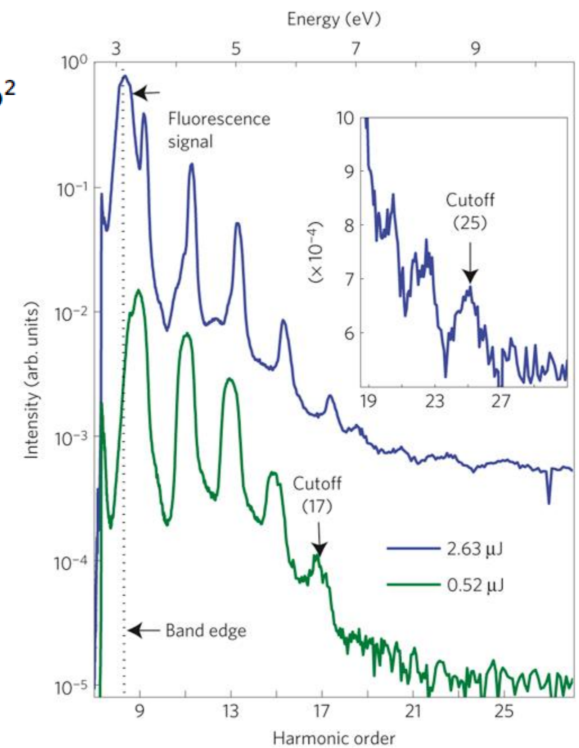


1. Tunneling   2. Acceleration by the field   3. Recombination

[1] Phys. Rev. Lett. 70, 1599 (1993); [2] Phys. Rev. Lett. 71, 1994 (1993)

And 30 years later… HHG in solids

Bulk ZnO



High-harmonic generation (HHG) in solids

LETTERS

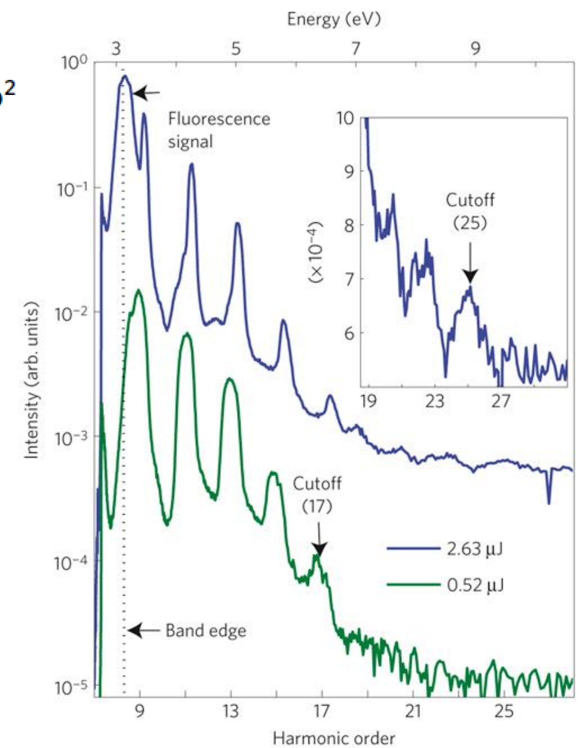
PUBLISHED ONLINE: 5 DECEMBER 2010 | DOI: 10.1038/NPHYS1847

nature physics

Observation of high-order harmonic generation in a bulk crystal

Shambhu Ghimire[1], Anthony D. DiChiara[2], Emily Sistrunk[2], Pierre Agostini[2], Louis F. DiMauro[2] and David A. Reis[1,3]*

Bulk ZnO



Cox *et al.*, Nat. Comm. 8, 14380 (2017)
Hammond *et al.*, Nat. Phot. 11, 594 (2017)
Langer *et al.*, Nat. Phot. (2017)
Liang *et al.*, Nat. Comm. 8 (2017)
Sivis *et al.*, Science 357,Nat. Comm. 303(2017)
Tancogne-Dejean *et al.*, 8, 745 (2017)
Vampa *et al.*, Nat. Phys. (2017)
You *et al.*, Nat. Comm. 8, 724 (2017)
Yoshikawa *et al.*, Science 356, 736(2017)

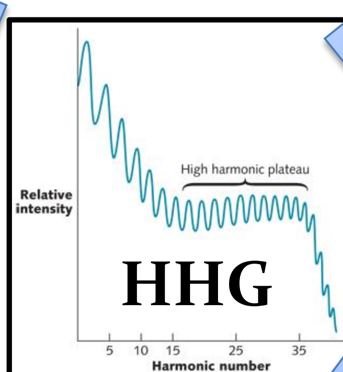
Some applications of HHG in solids

Micrometer-scale extreme-ultra-violet (XUV) sources

Table-top synchrotron

Electron-hole recollisions in real time

Zaks *et al* Nature 483, 580 (2012).



All-optical band-structure reconstruction

Vampa *et al.*, PRL. 115, 193603 (2015).

Quantum-logic at optical clock-rates

Understanding HHG in solids

What is the microscopic mechanism responsible for HHG in solids?

Understanding HHG in solids

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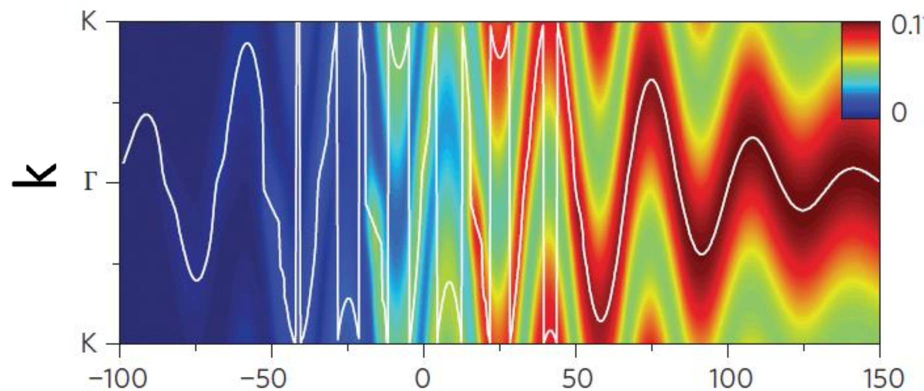
What is the microscopic mechanism responsible for HHG in solids?

Dynamical Bloch oscillations?



From Schubert *et al.* Nature Photonics **8**, 119 (2014)

Understanding HHG in solids

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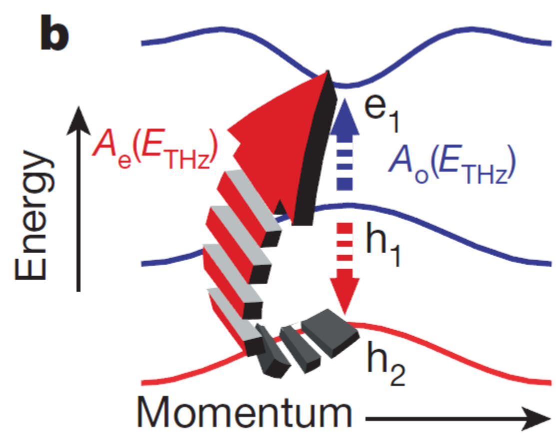
Dynamical Bloch oscillations?

Interband transitions?



From Hohenleutner *et al.* Nature **523**, 572 (2015)

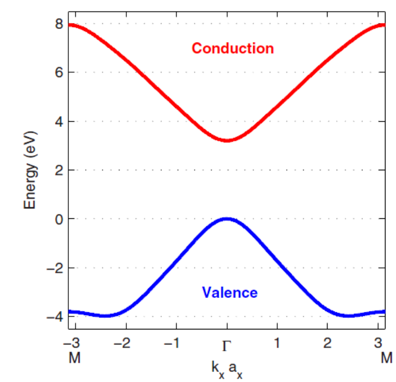
How many bands are contributing?

Understanding HHG in solids

What is the microscopic mechanism responsible for HHG in solids?

Interband transitions?

Two-band model?



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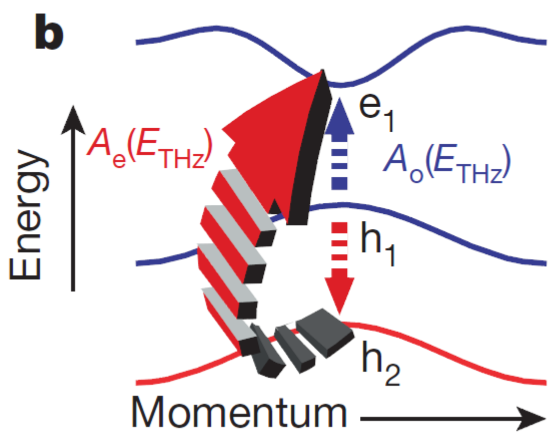
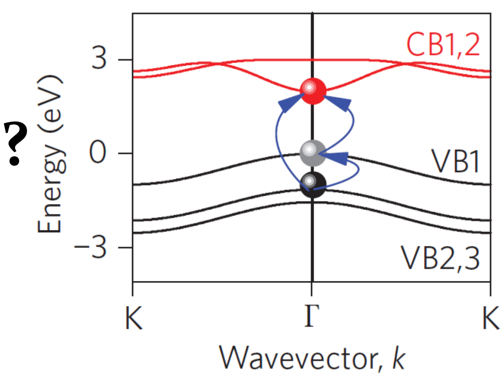


From Hohenleutner *et al.* Nature **523**, 572 (2015)

Five-band model?



From Schubert *et al.*
Nature Photonics **8**, 119 (2014)

Open questions

Which model should I use?

Open questions

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Are electrons independent particles, i.e., what is the role of correlations?

What is the role of the surface, phonons, light-propagation?

Which material should I use ?

Ab initio approach to HHG in solids

Time-dependent density functional theory (TDDFT) framework

- No empirical parameters
- Full band-structure included, real crystal structure
- No *a priori* approximation on the number of bands
- Correlation effects can be investigated
- Possibility to go beyond intrinsic effects:

Phonons and surface effects,
light propagation effects,
…

Ab initio approach to HHG in solids

TDDFT framework with **Octopus** code

- Dipole approximation
- Laser is modeled by a time-dependent vector potential
- Real-space real-time TDDFT

Some exact analytical results

Let us consider a general Hamiltonian

$$\hat{H}(t) = \hat{T} + \hat{V}(t) + \hat{W},$$

From the equation of motion of the electronic current

$$\frac{\partial}{\partial t}\mathbf{j}(\mathbf{r},t) = -i\langle\Psi(t)|[\hat{\mathbf{j}}(\mathbf{r}),\hat{H}(t)]|\Psi(t)\rangle$$

$$\frac{\partial}{\partial t}\mathbf{j}(\mathbf{r},t) = -n(\mathbf{r},t)\nabla v(\mathbf{r},t) + \Pi^{\text{kin}}(\mathbf{r},t) + \Pi^{\text{int}}(\mathbf{r},t)$$

[1] N. T-D *et al.*,PRL 118, 087403 (2017)

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Momentum of the system

Internal forces of the system

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Momentum of the system

Internal forces of the system

Third Newton's law: only external forces contribute to the total momentum of the system

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we obtain

$$\frac{\partial}{\partial t}\int_{\Omega} d^3\mathbf{r}\,\mathbf{j}(\mathbf{r},t) = -\int_{\Omega} d^3\mathbf{r}\,n(\mathbf{r},t)\nabla v(\mathbf{r},t).$$

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$$\boxed{\mathrm{HHG}(\omega) = \left|\mathrm{FT}\left(\frac{\partial}{\partial t}\int d^3\mathbf{r}\,\mathbf{j}(\mathbf{r},t)\right)\right|^2}$$

[1] N. T-D *et al.*,PRL 118, 087403 (2017)

Some exact analytical results

From the *exact* equation of motion of the electronic current, we can write that [1]

$$\mathrm{HHG}(\omega) \propto \left| \mathrm{FT}\left(\int_\Omega d^3\mathbf{r}\, n(\mathbf{r},t) \nabla v_0(\mathbf{r}) \right) + N_e \mathbf{E}(\omega) \right|^2$$

Valid for atom, molecules and solids (dipole approximation)

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Valid for atom, molecules and solids (dipole approximation)

- HHG originate from competing terms: electronic density and electron-ion potential
- No HHG from an homogeneous electron gas (parabolic bands)
- HHG is enhanced by inhomogeneity of the electron-ion potential -> layered materials are good candidates for HHG

[1] N. T-D *et al.*,PRL 118, 087403 (2017)

Correlation effects in HHG

What is the role of correlations in HHG in solids?

Time-dependent Kohn-Sham equations

$$i\frac{\partial}{\partial t}\phi_i(\mathbf{r},t) = \left(-\frac{\nabla^2}{2} + v_{\rm ext}(\mathbf{r},t) + v_{\rm H}[n](\mathbf{r},t) + v_{\rm xc}[n](\mathbf{r},t)\right)\phi_i(\mathbf{r},t)$$

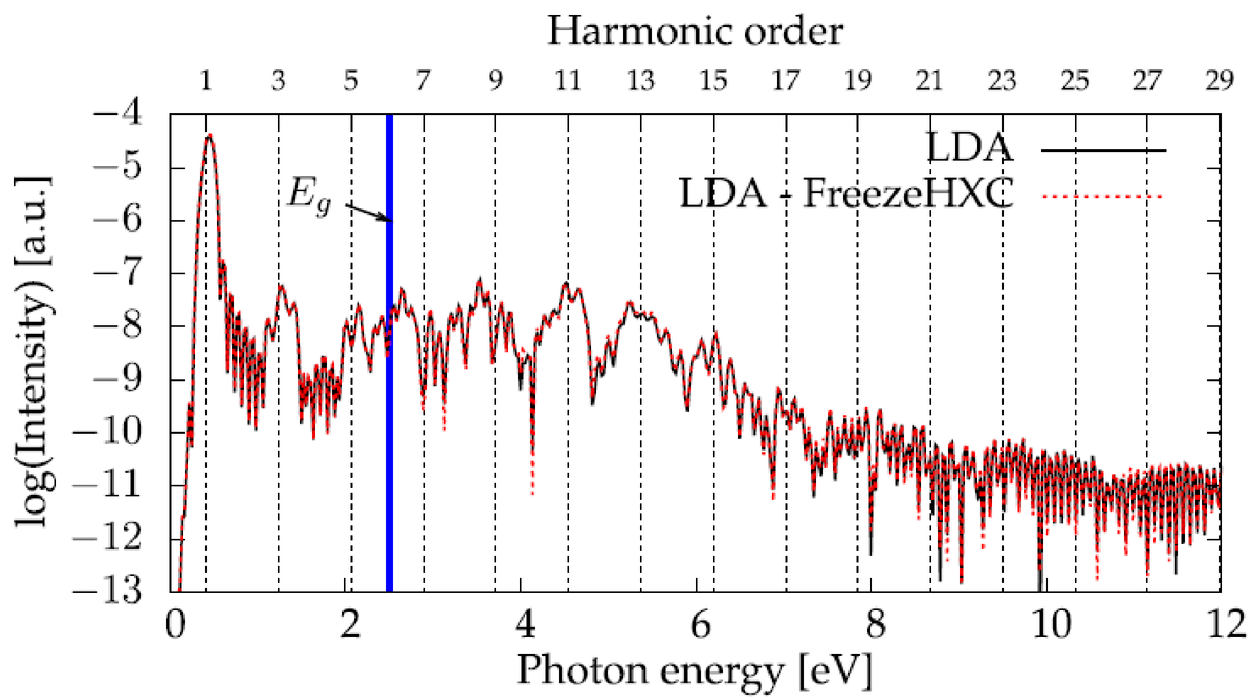
Independent-particle approximation:

$$i\frac{\partial}{\partial t}\phi_i(\mathbf{r},t) = \left(-\frac{\nabla^2}{2} + v_{\rm ext}(\mathbf{r},t) + v_{\rm H}[\underline{n_0}](\mathbf{r}) + v_{\rm xc}[\underline{n_0}](\mathbf{r})\right)\phi_i(\mathbf{r},t)$$

Correlation effects in HHG

mpsd

In bulk silicon, the Hartree and exchange-correlation potentials do not evolve during the laser pulse.

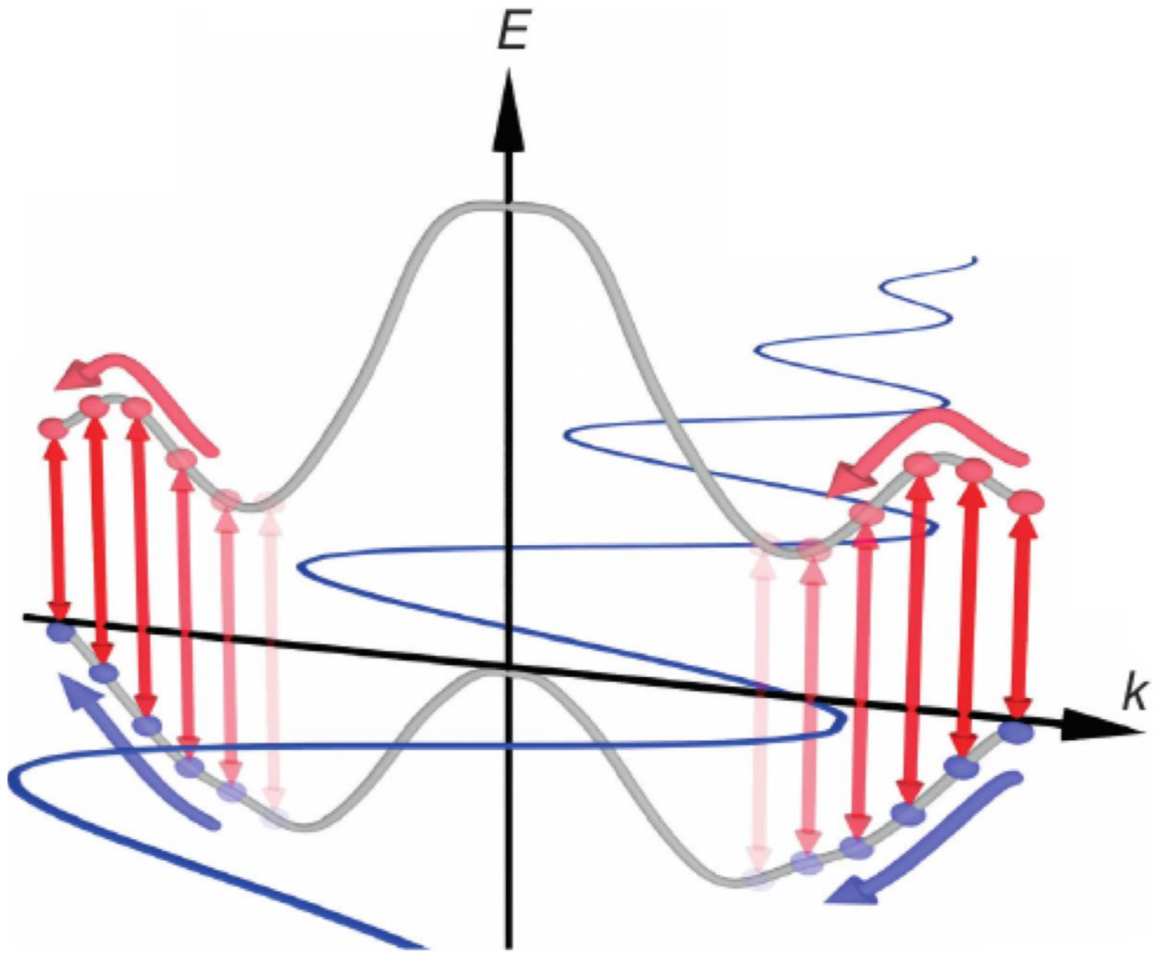


- ➢Electrons evolve in a fixed band structure
- ➢Band structure might be retrieved

Bulk Silicon
λ=3000nm
25fs FWHM
I=3.4×10^{11} W/cm^2

[1] N. T-D *et al.*,PRL 118, 087403 (2017)

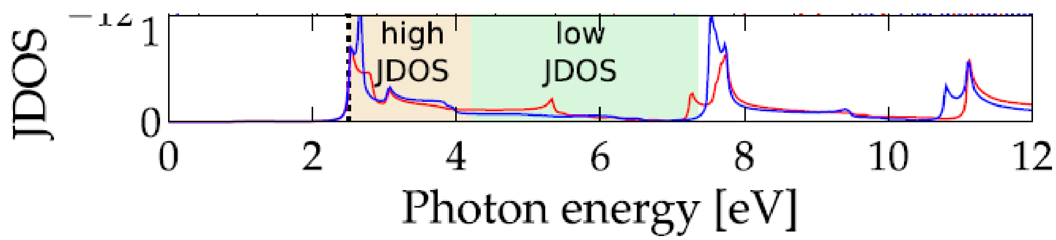
Interband *vs* Intraband mechanism



Adapted from Langer et al., Nature 533, 225 (2016)

Interband *vs* Intraband mechanism

Harmonic emission from interband mechanism:
only if conduction-valence transitions are available
The interband mechanism depends on the *density of optical transitions* (JDOS)



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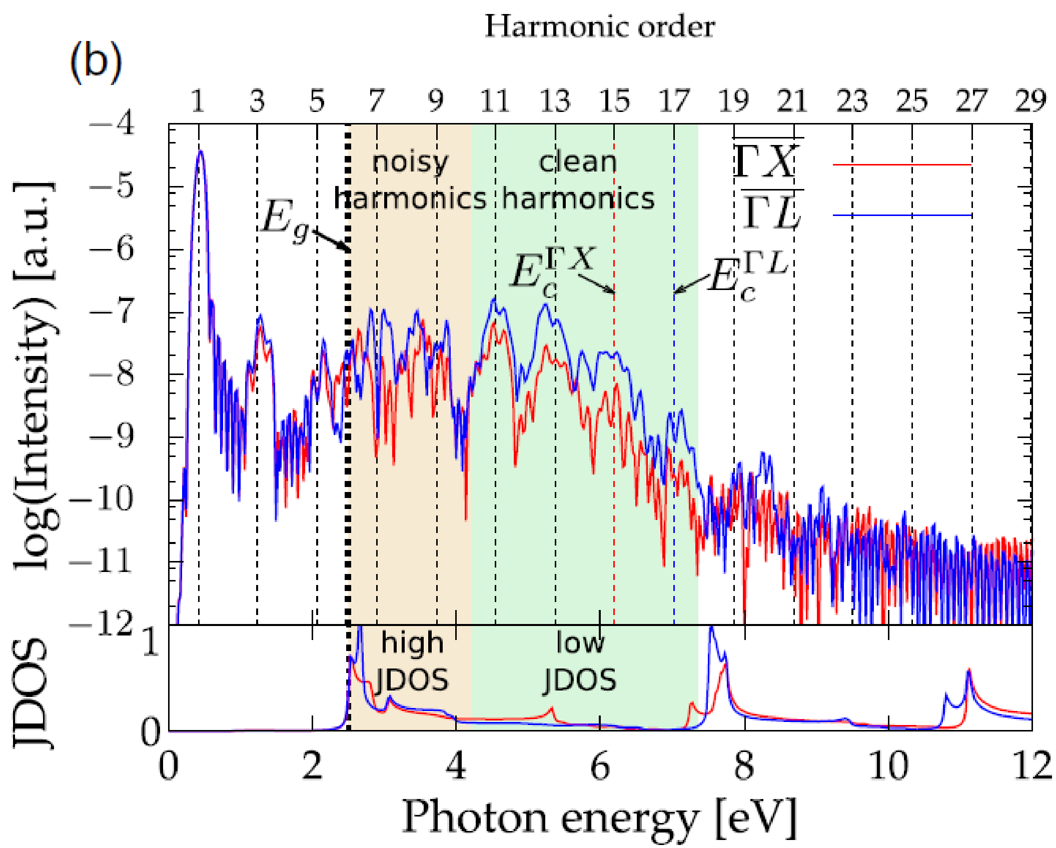
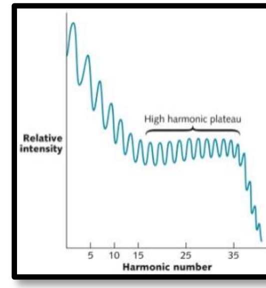


- Low JDOS: interband contribution is suppressed
- HHG yield improved when interband is suppressed
- Toward band-structure engineering to improve HHG in solids

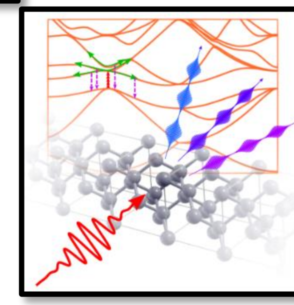
[1] N. T-D *et al.*,PRL 118, 087403 (2017)

Outline

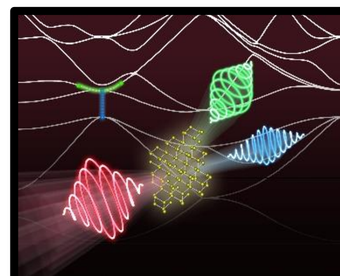
- High-harmonic generation (HHG)



- Impact of the band-structure



- **Ellipticity dependence**



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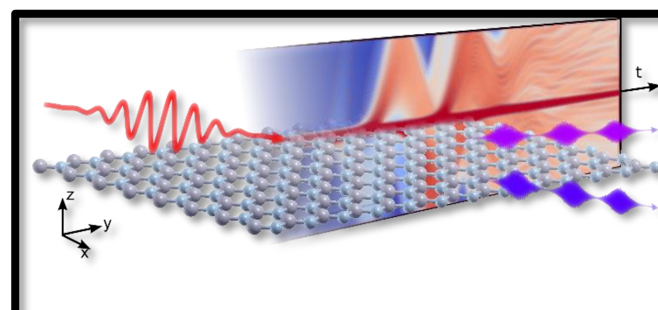
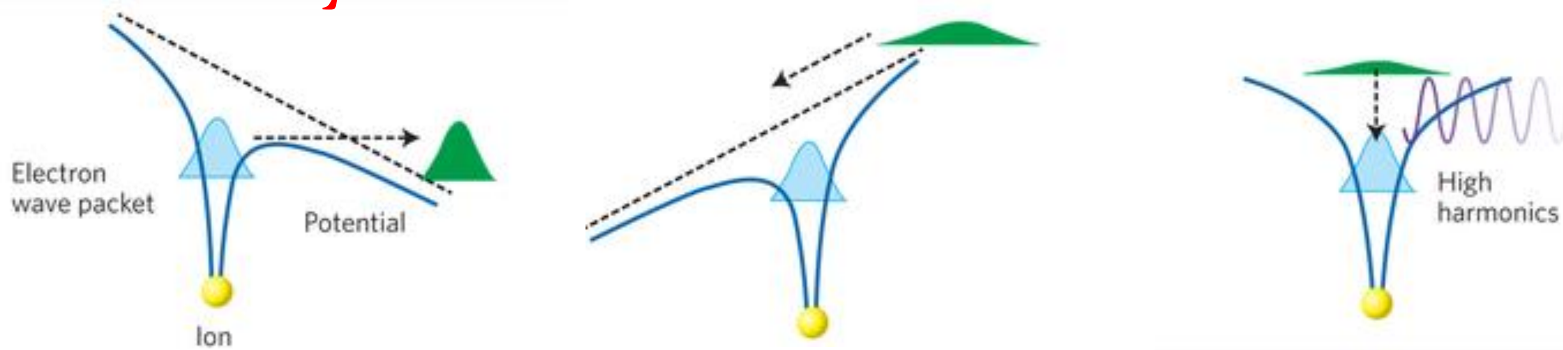
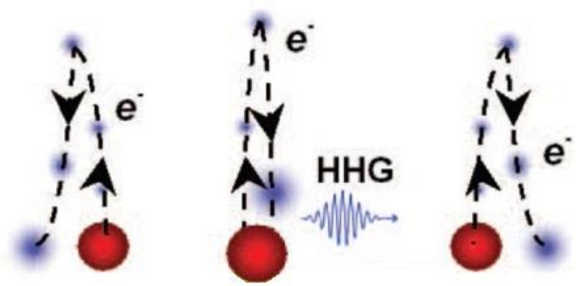


Ellipticity dependence in gases

In atomic gases, circular light suppresses the harmonic yield



Electrons acquire a transversal momentum and "misses" the parent ion.
No recombination, no harmonic emission



B. Shan *et al.*, J. Mod. Opt. **52**, 277 (2005)

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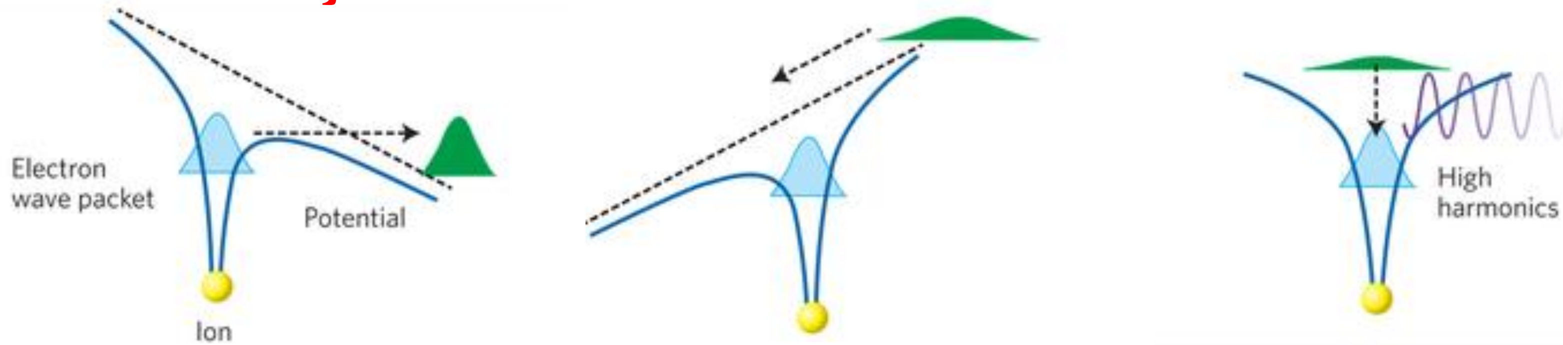

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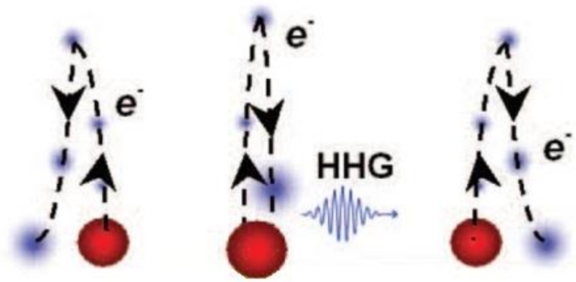

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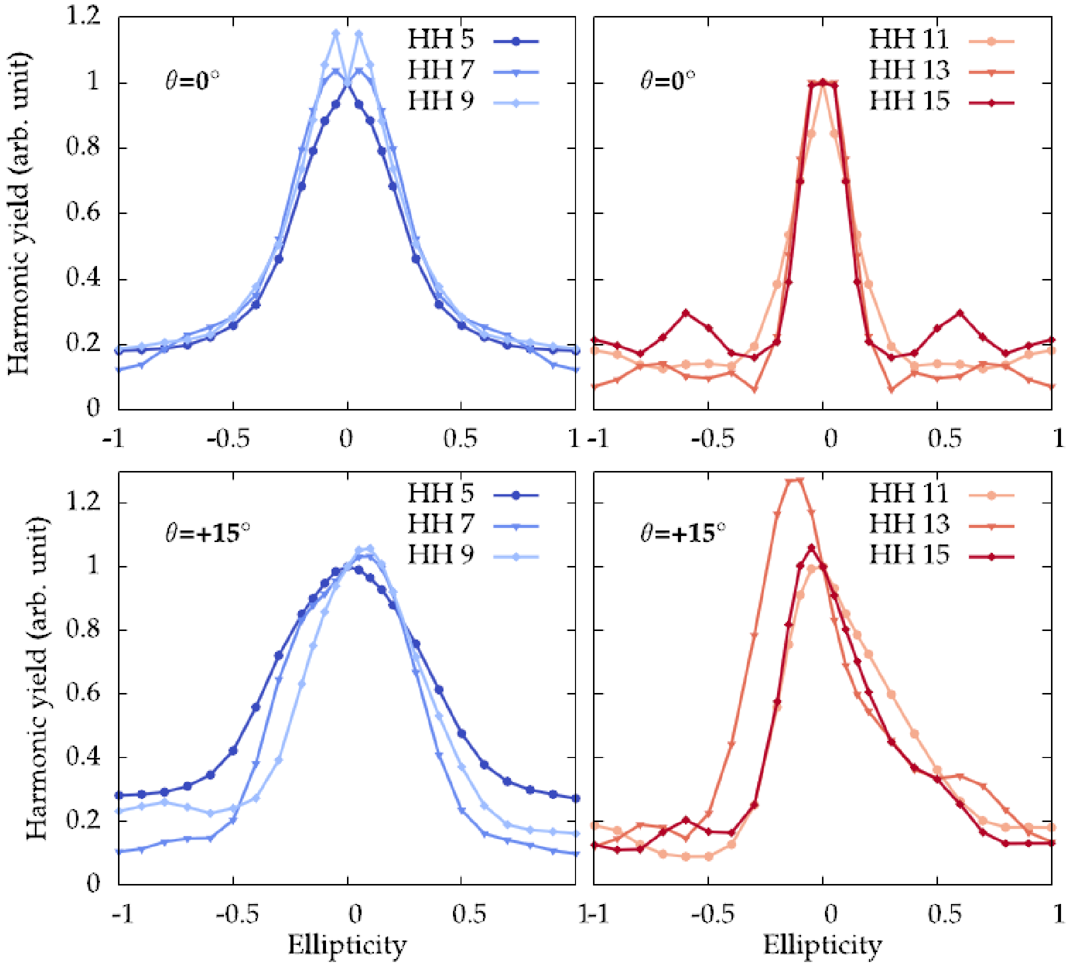
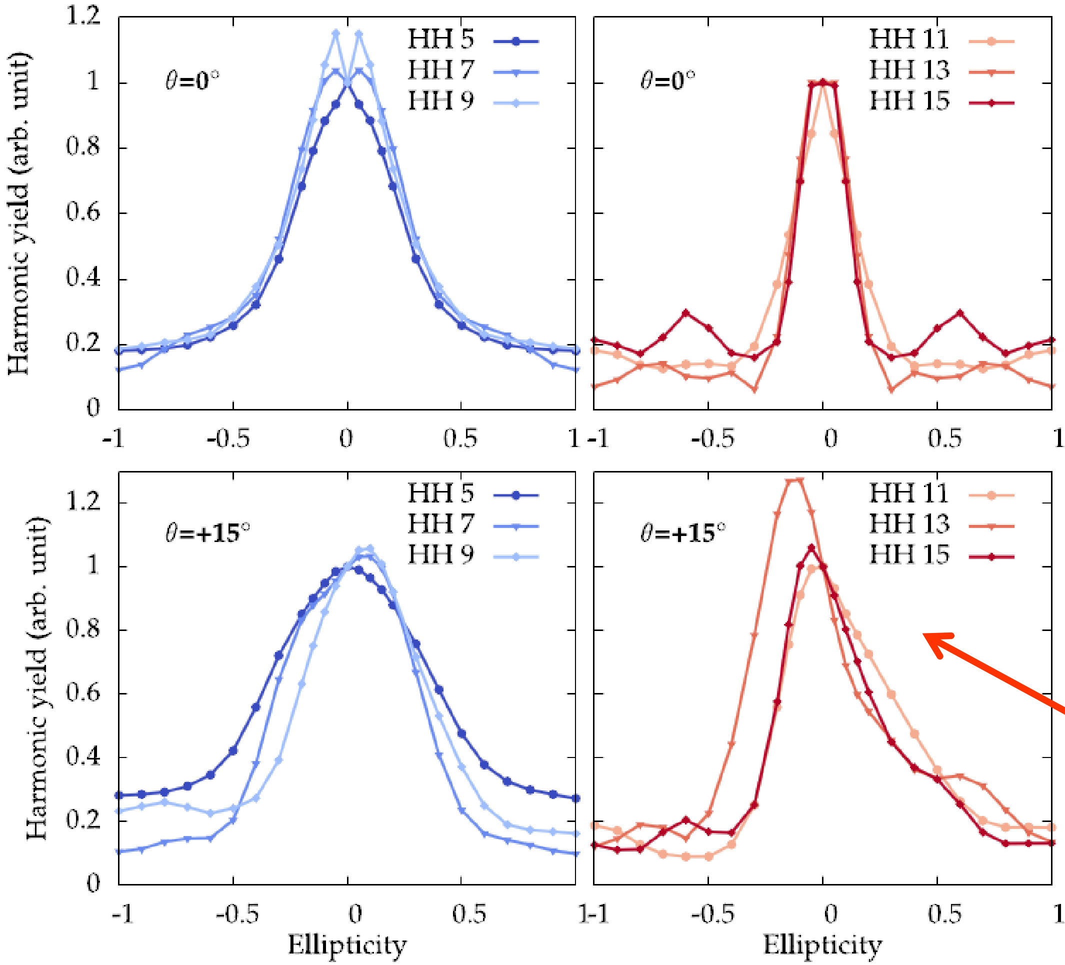
Not the case in solids !

Ellipticity dependence of HHG in solids



Bulk Si
λ=3000nm
I=3x10^{12} W/cm^2
25fs FWHM

Ellipticity dependence of HHG in solids



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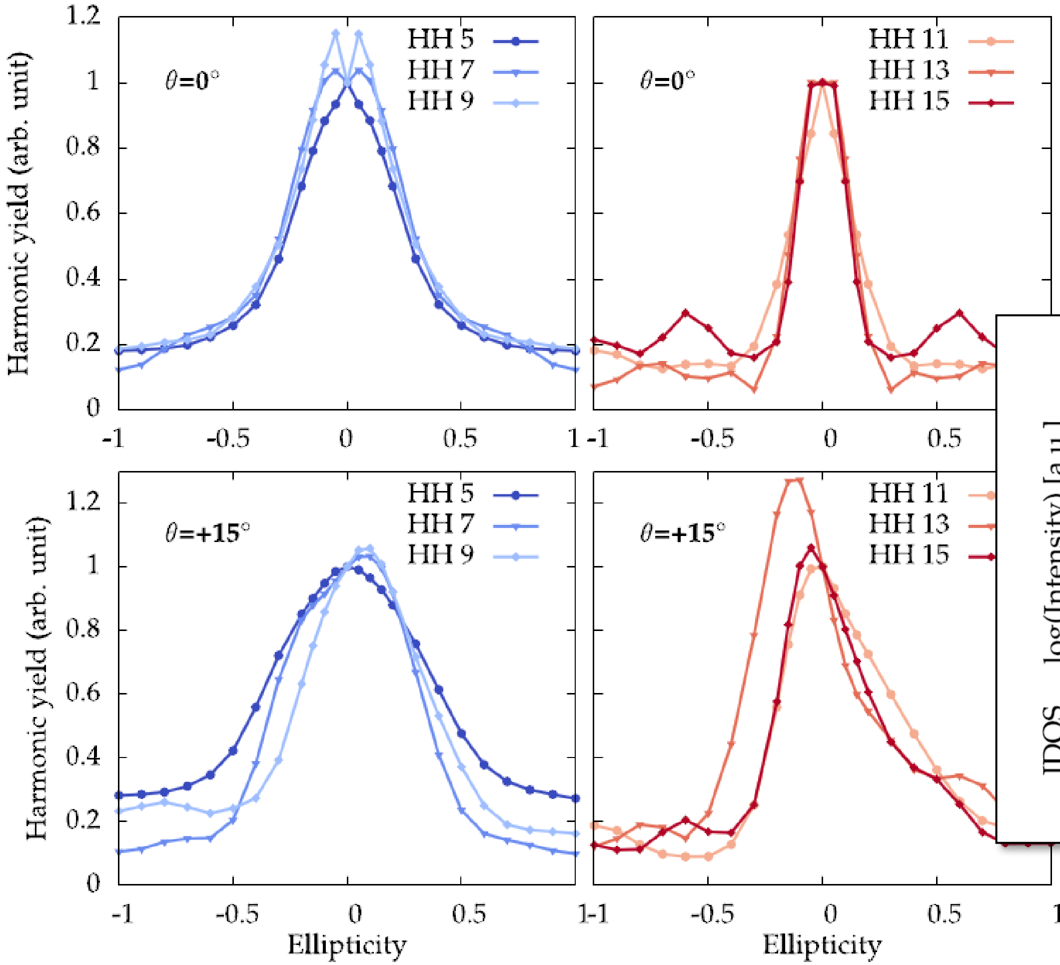
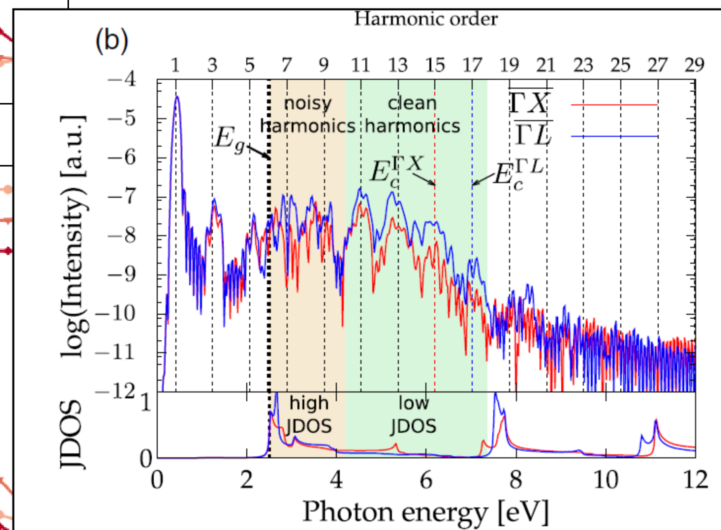
Anisotropic ellipticity profiles

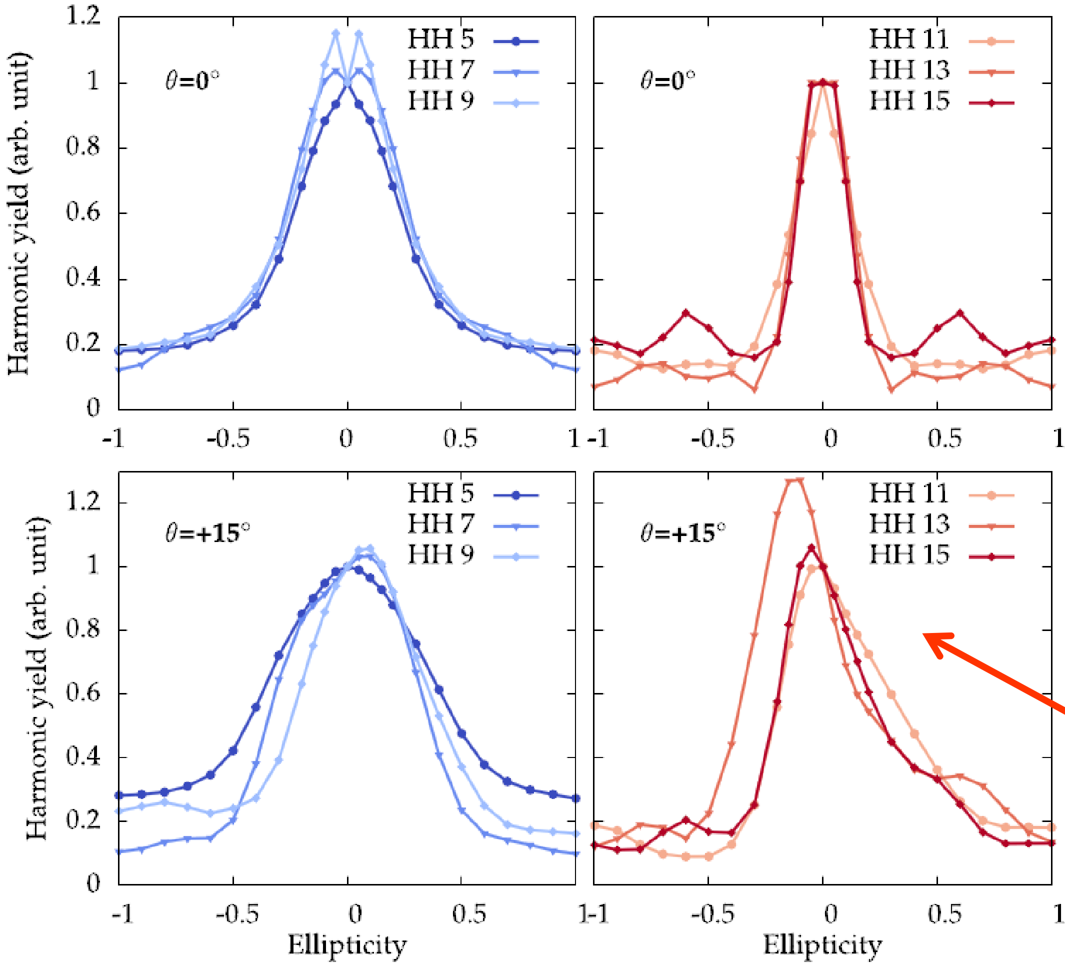
Ellipticity dependence of HHG in solids





Bulk Si
λ=3000nm
I=3x10[12] W/cm[2]
25fs FWHM

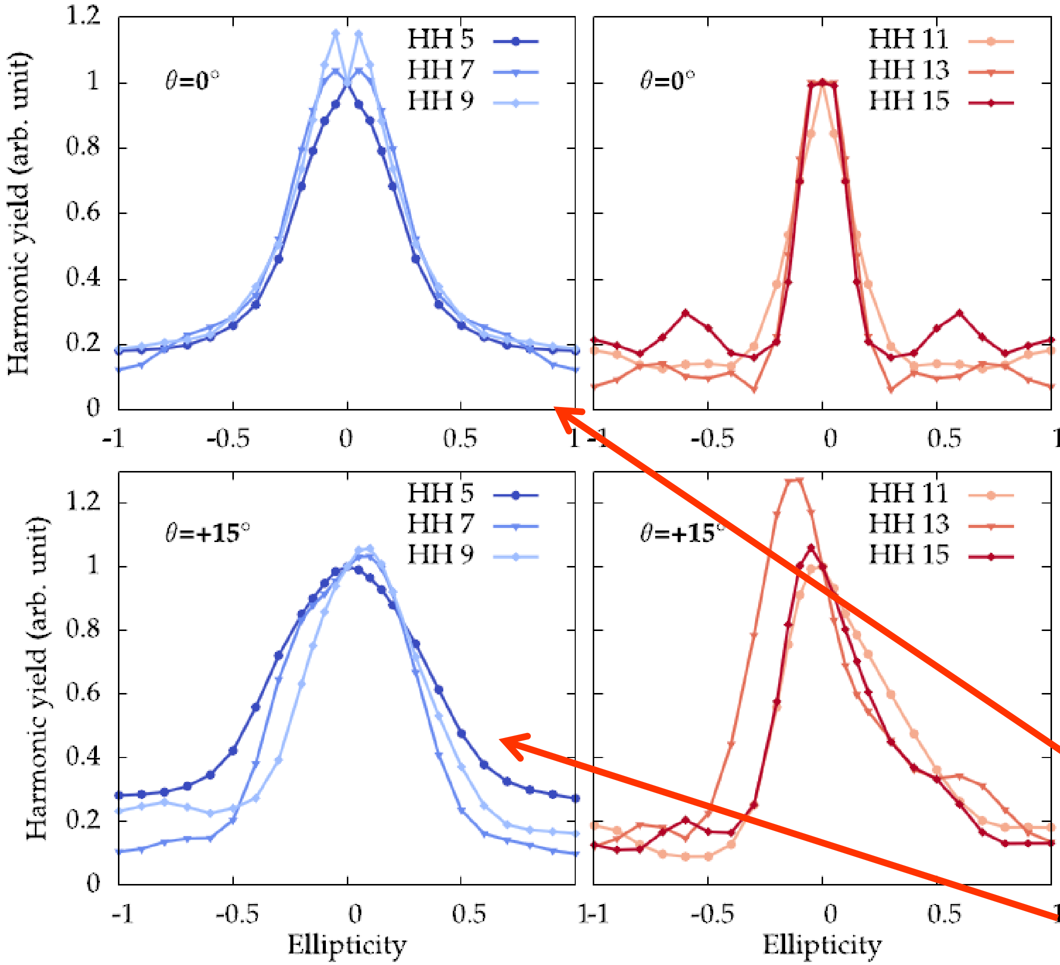
Ellipticity dependence of HHG in solids



Bulk Si
λ=3000nm
I=3x10^{12} W/cm^2
25fs FWHM

Mostly from intraband

Ellipticity dependence of HHG in solids

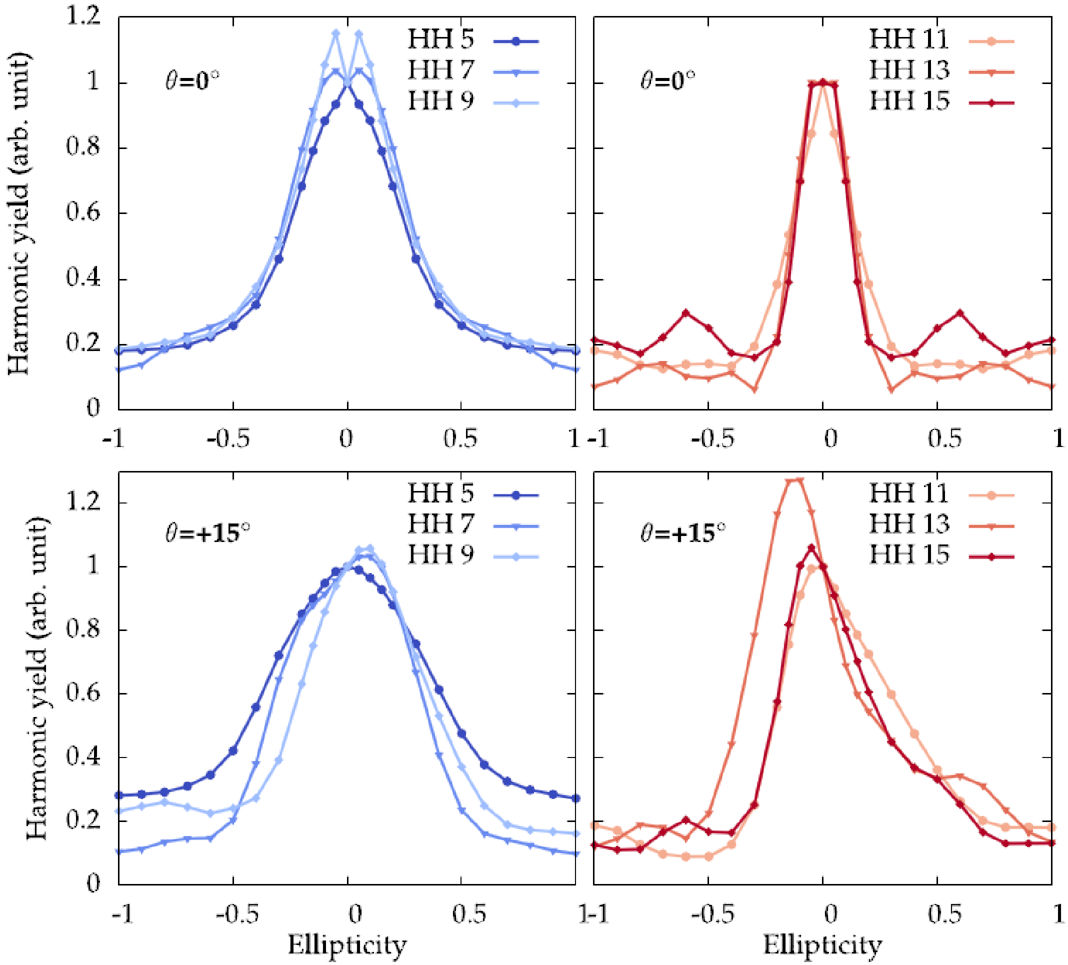


Bulk Si
λ=3000nm
I=3x10^{12} W/cm^2
25fs FWHM

Interband+ intraband

Ellipticity dependence of HHG in solids

mpsd

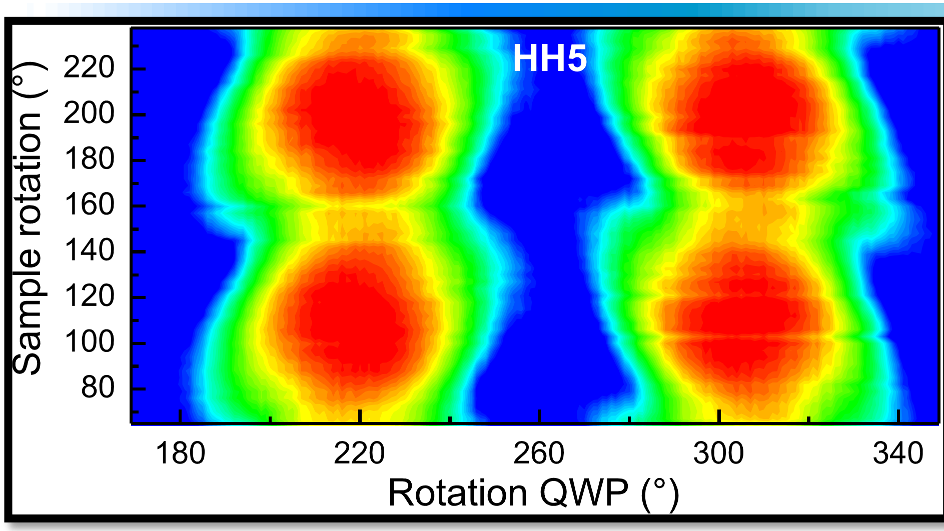


Bulk Si
λ=3000nm
$I=3x10^{12}$ W/cm²
25fs FWHM

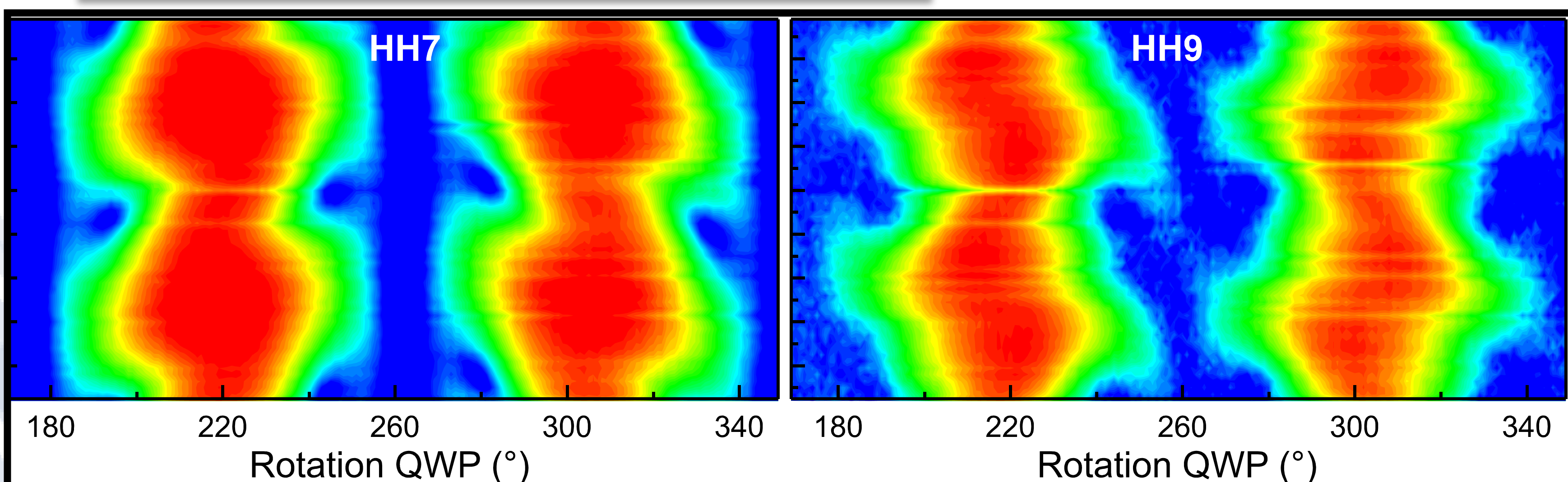
Interband and intraband react differently the driver ellipticity

[1] N. T.-D. *et al.* Nature Comm. 8, 745 (2017)

Ellipticity dependence of HHG in solids

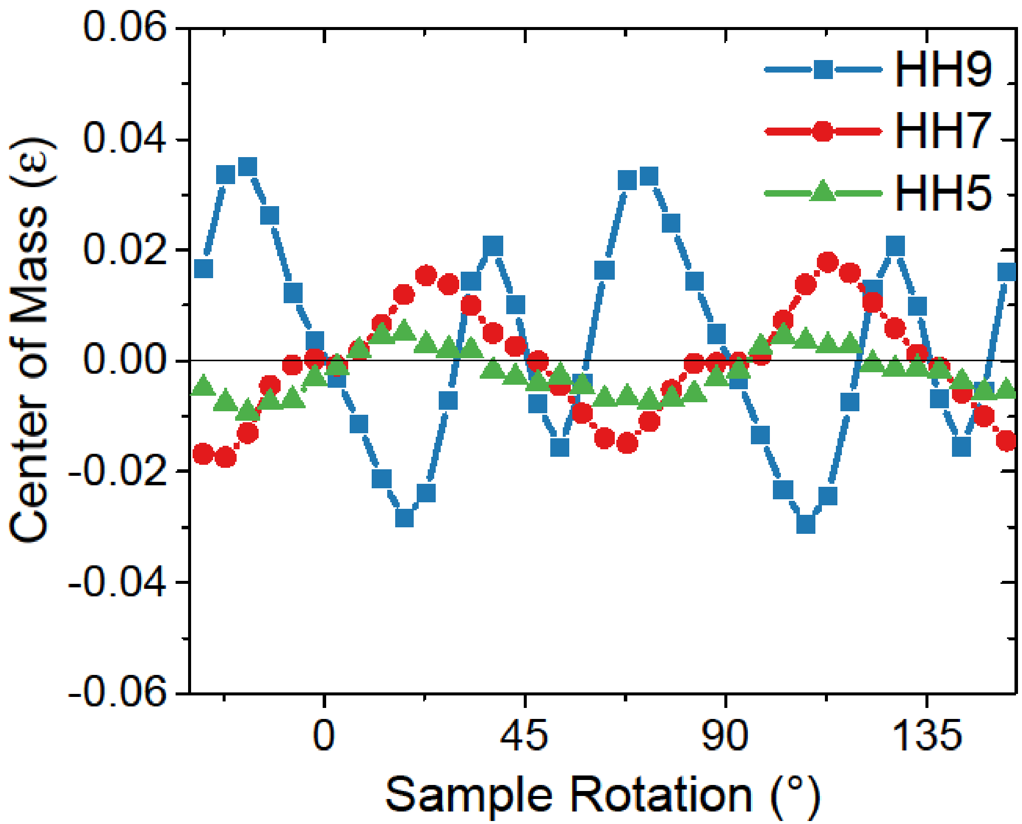


Free-standing 2μm Si(001)
λ=2.1 μm
I=0.6 TW/cm² in vacuum
70fs FWHM



N. Klemke *et al.*, Polarization-state-resolved high-harmonic spectroscopy of solids (Submitted)

Experimental evidence



Free-standing 2µm Si(001)
λ=2.1 µm
I=0.6 TW/cm² in vacuum
70fs FWHM

HH7 and HH9 oscillate in opposite directions

N. Klemke *et al.*, *Polarization-state-resolved high-harmonic spectroscopy of solids* (Submitted)

Circularly polarized harmonics from solids

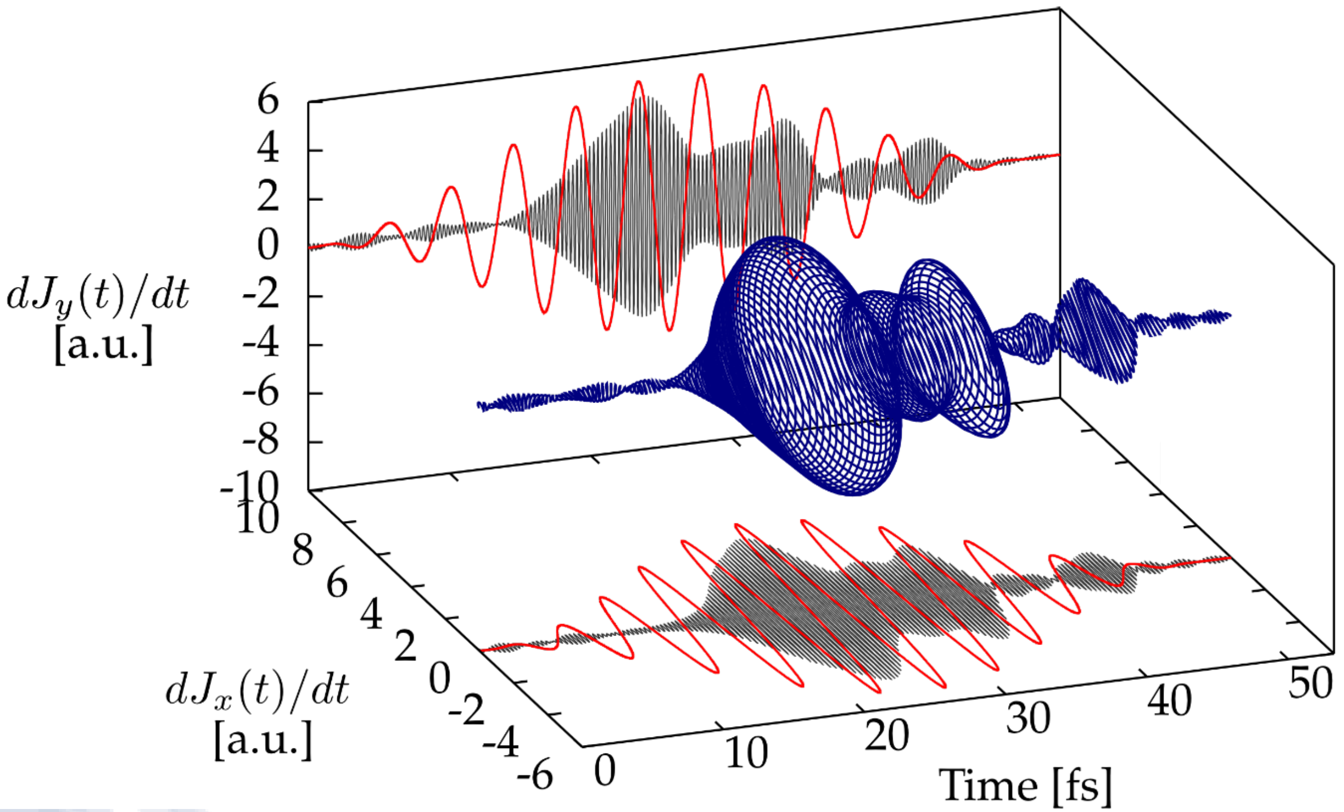
What about the emitted harmonics?

Circularly polarized harmonics from solids

Emitted harmonics follow the ellipticity of the driver field

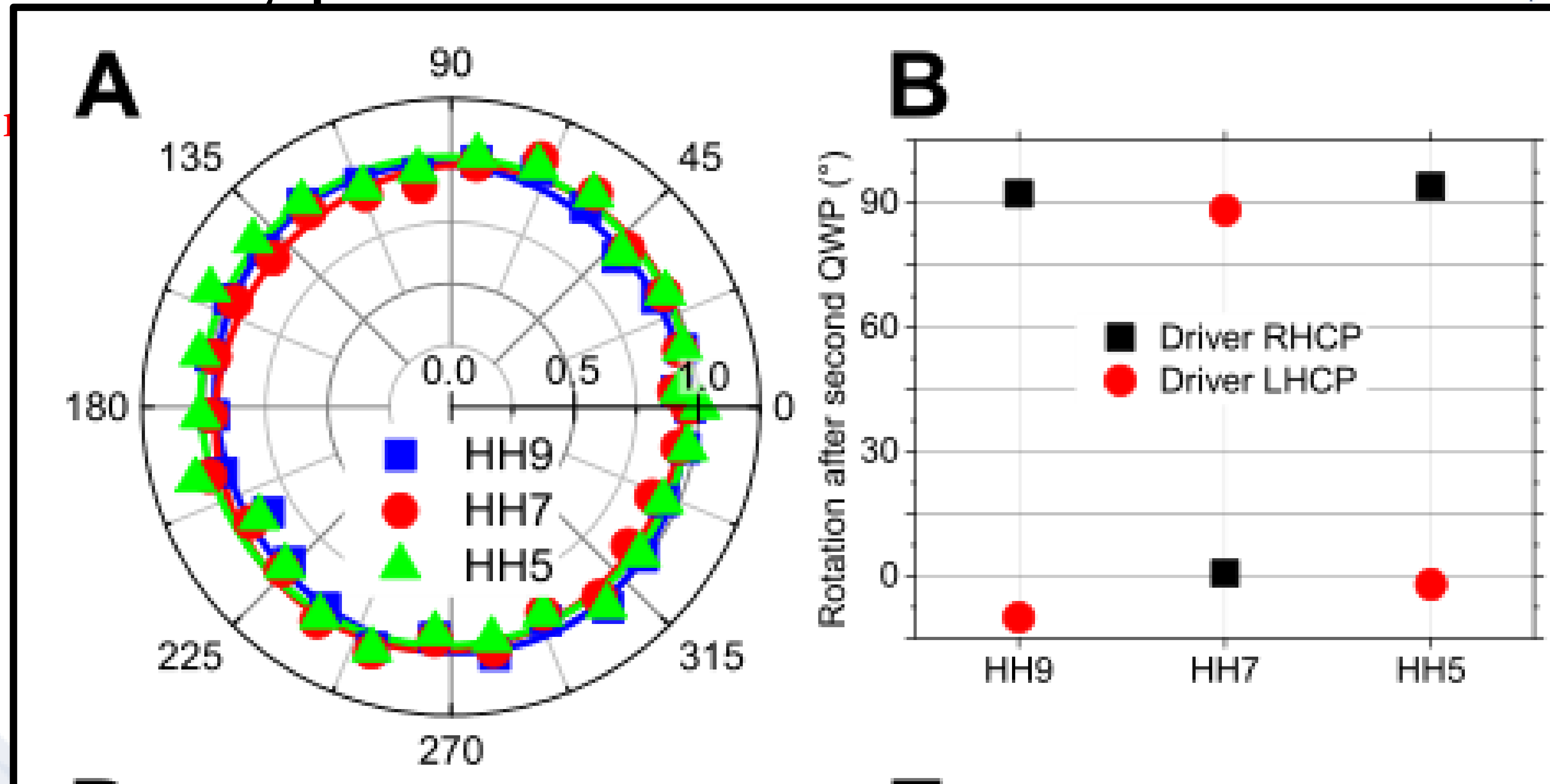
Example: Harmonic 15th in MgO

λ=1333nm,
I=3x10^{12} W/cm^2
50fs FWHM



[1] N. T.-D. *et al.* Nature Comm. 8, 745 (2017)

Circularly polarized harmonics from solids



N. Klemke et al.

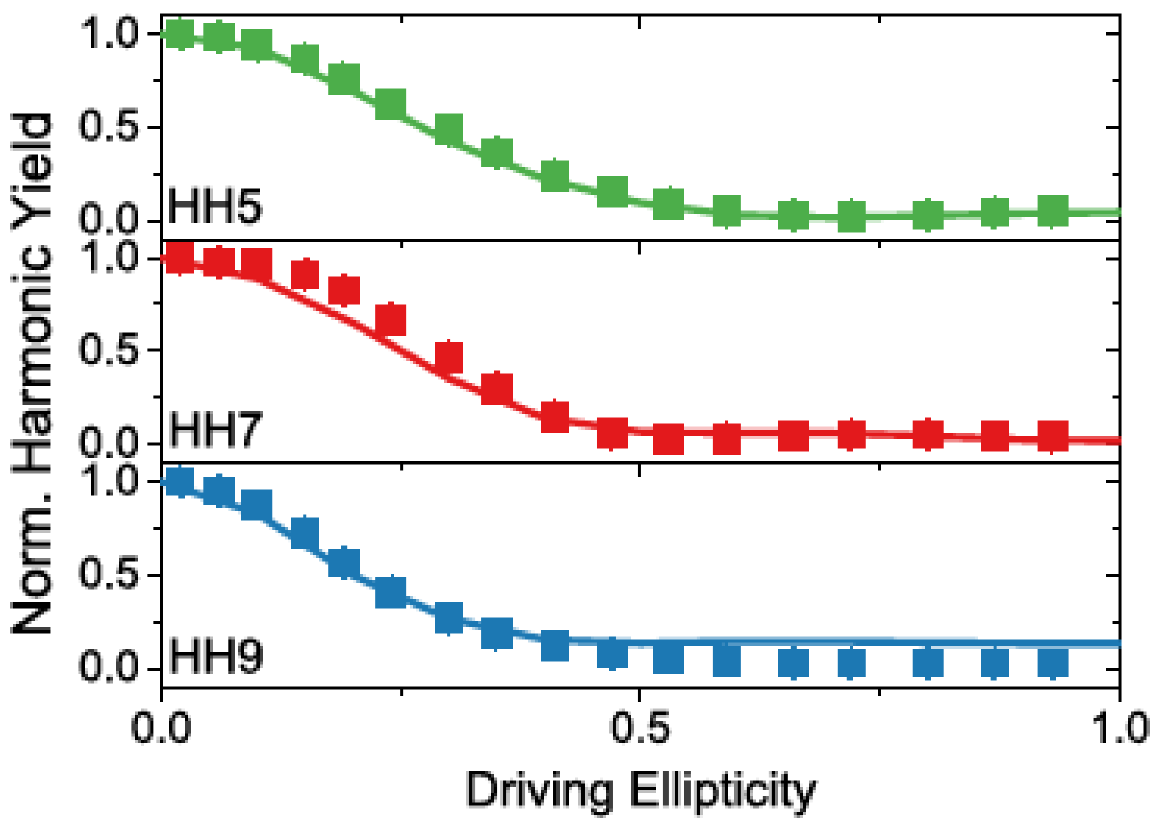
Polarization-state-resolved high-harmonic spectroscopy of solids

(Submitted)

from coupled intraband and interband dynamics Nature Comm. 8, 745 (2017)

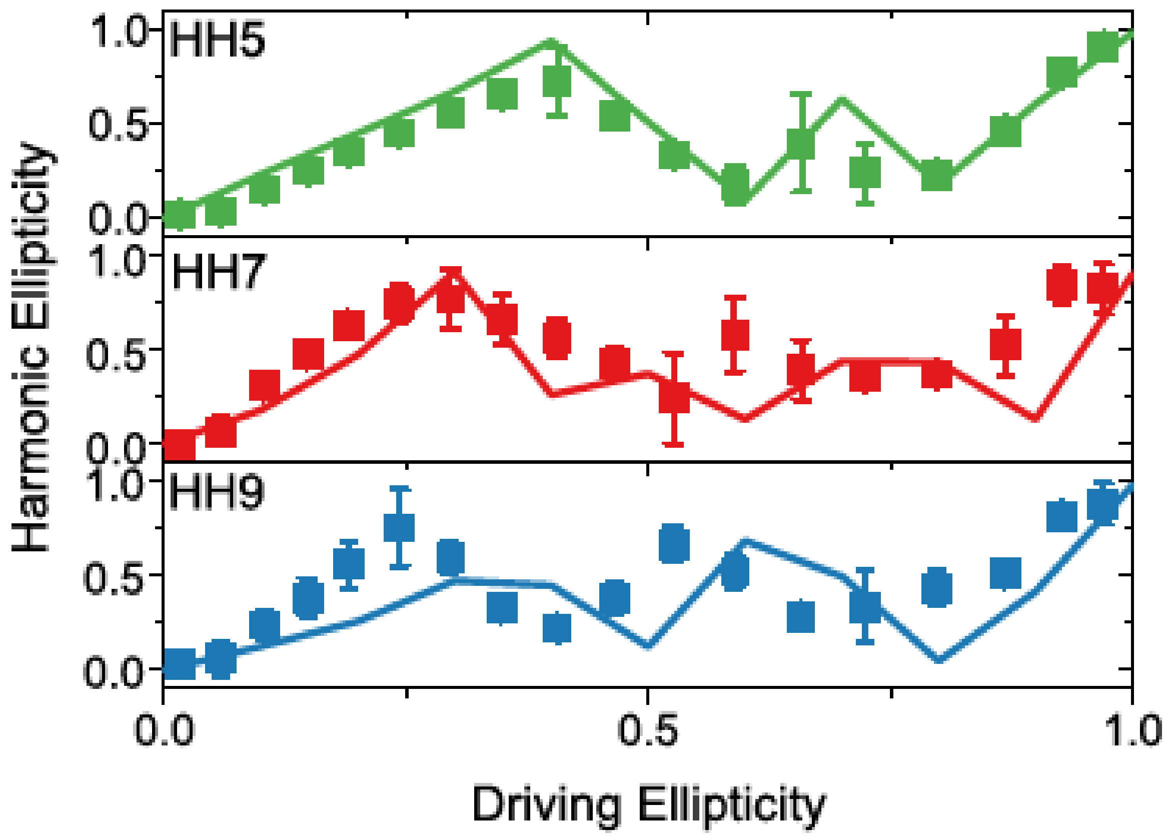
Does it work ?

Polarization states of the harmonics: theory vs exp.



N. Klemke *et al., Polarization-state-resolved high-harmonic spectroscopy of solids* (Submitted)

Polarization states of the harmonics: theory vs exp.



N. Klemke *et al.*, *Polarization-state-resolved high-harmonic spectroscopy of solids* (Submitted)

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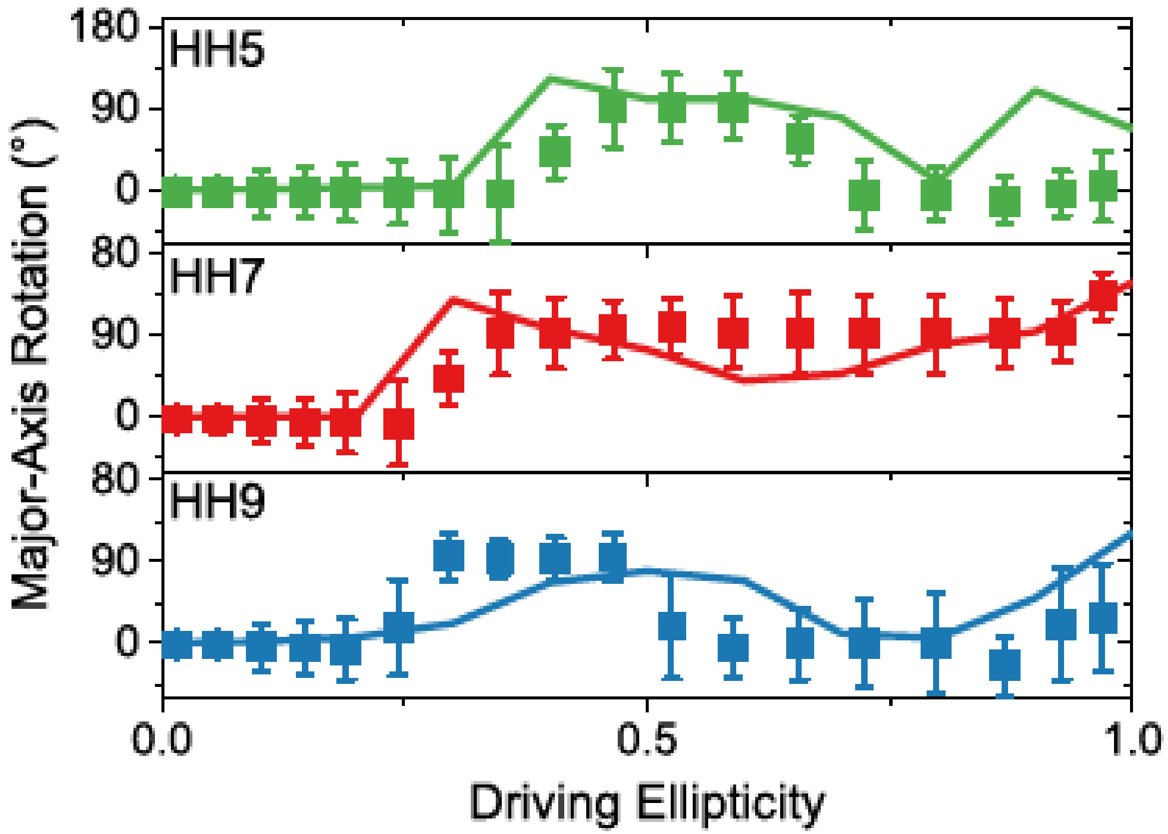
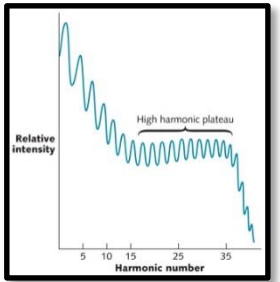


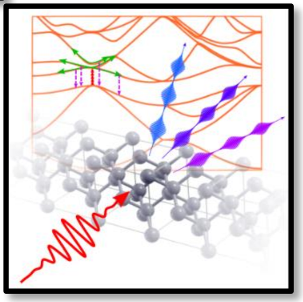
N. Klemke *et al.*, *Polarization-state-resolved high-harmonic spectroscopy of solids* (Submitted)

Outline

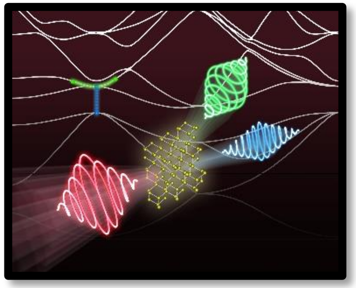
- High-harmonic generation (HHG)



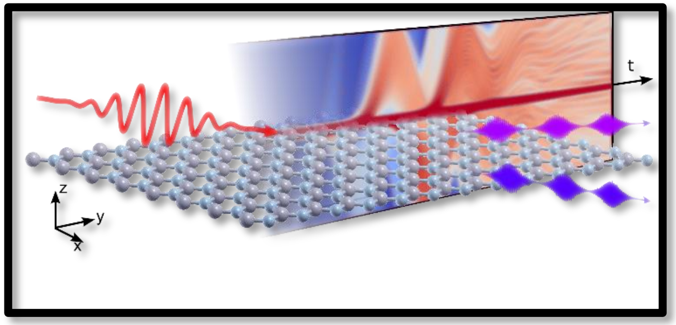
- Impact of the band-structure



- Ellipticity dependence



- Atomic-like HHG from 2D materials

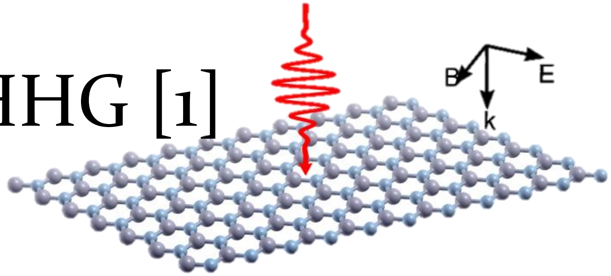


HHG from monolayer materials

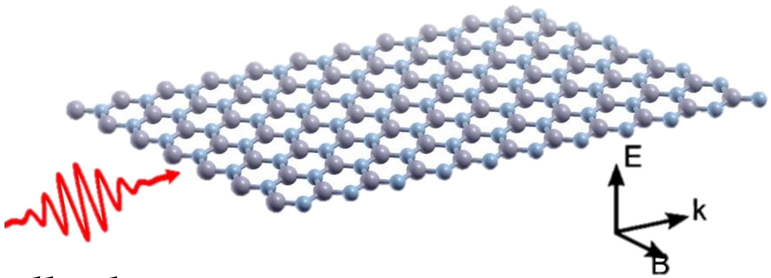
What happens in quasi-two-dimensional materials?

Two cases:

- in-plane electric field -> bulk-like HHG [1]

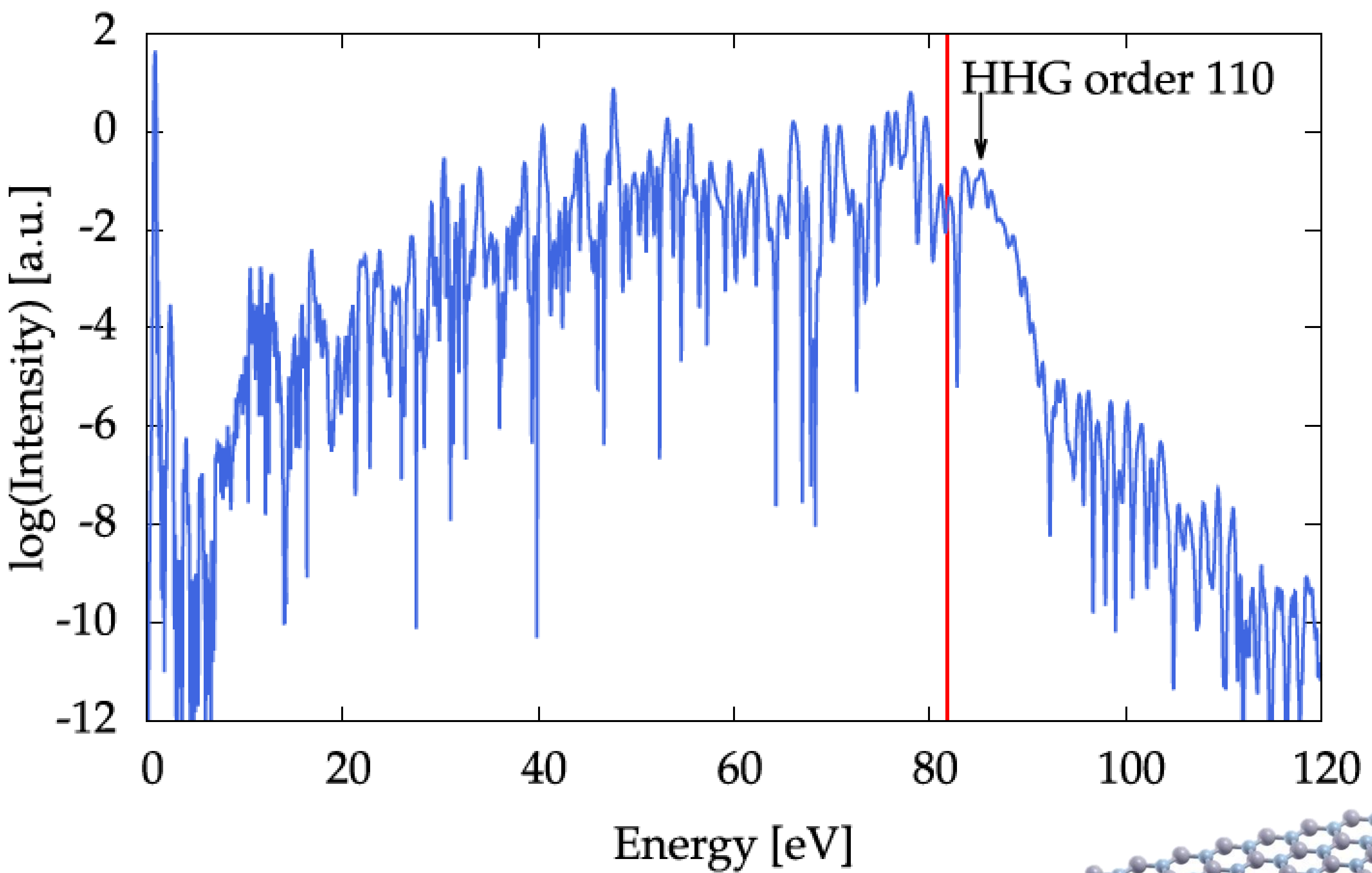


- out-of-plane electric field -> atomic-like HHG ?



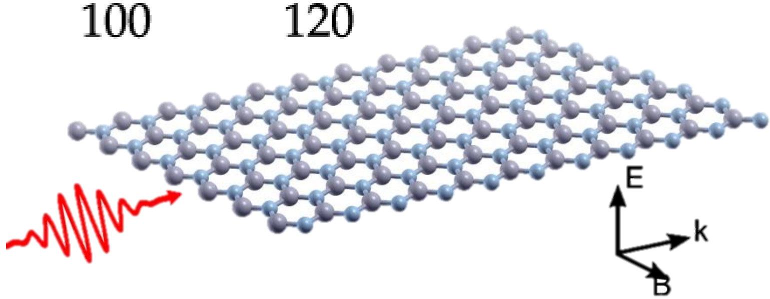
[1] Liu *et al. High-harmonic generation from atomically thin semiconductor.* Nature Physics (2016)

Atomic-like HHG from 2D materials

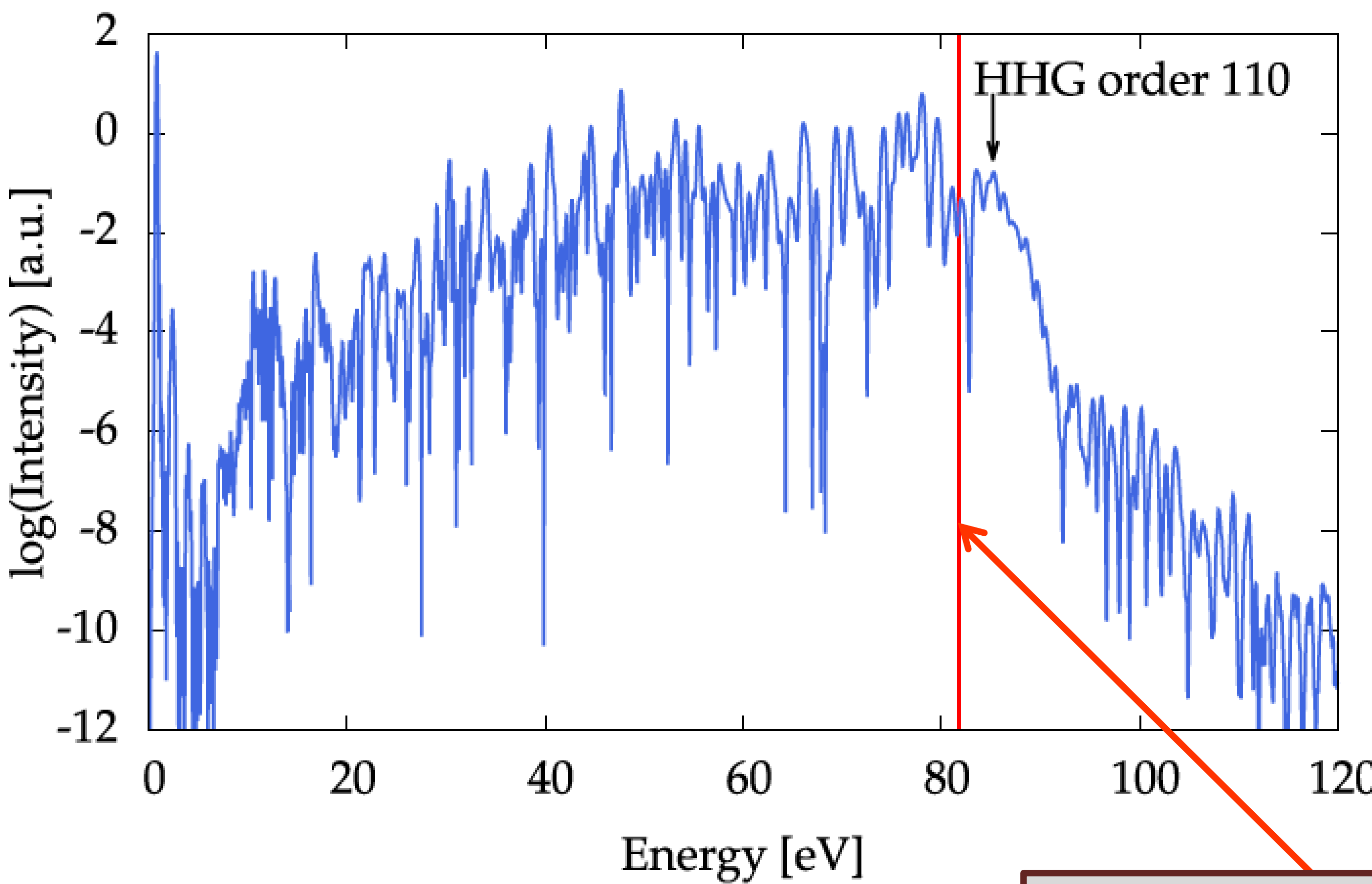


hBN
λ=1600nm
I=1x10^{14} W/cm^2
15fs FWHM

K-point grid
42x42



Atomic-like HHG from 2D materials



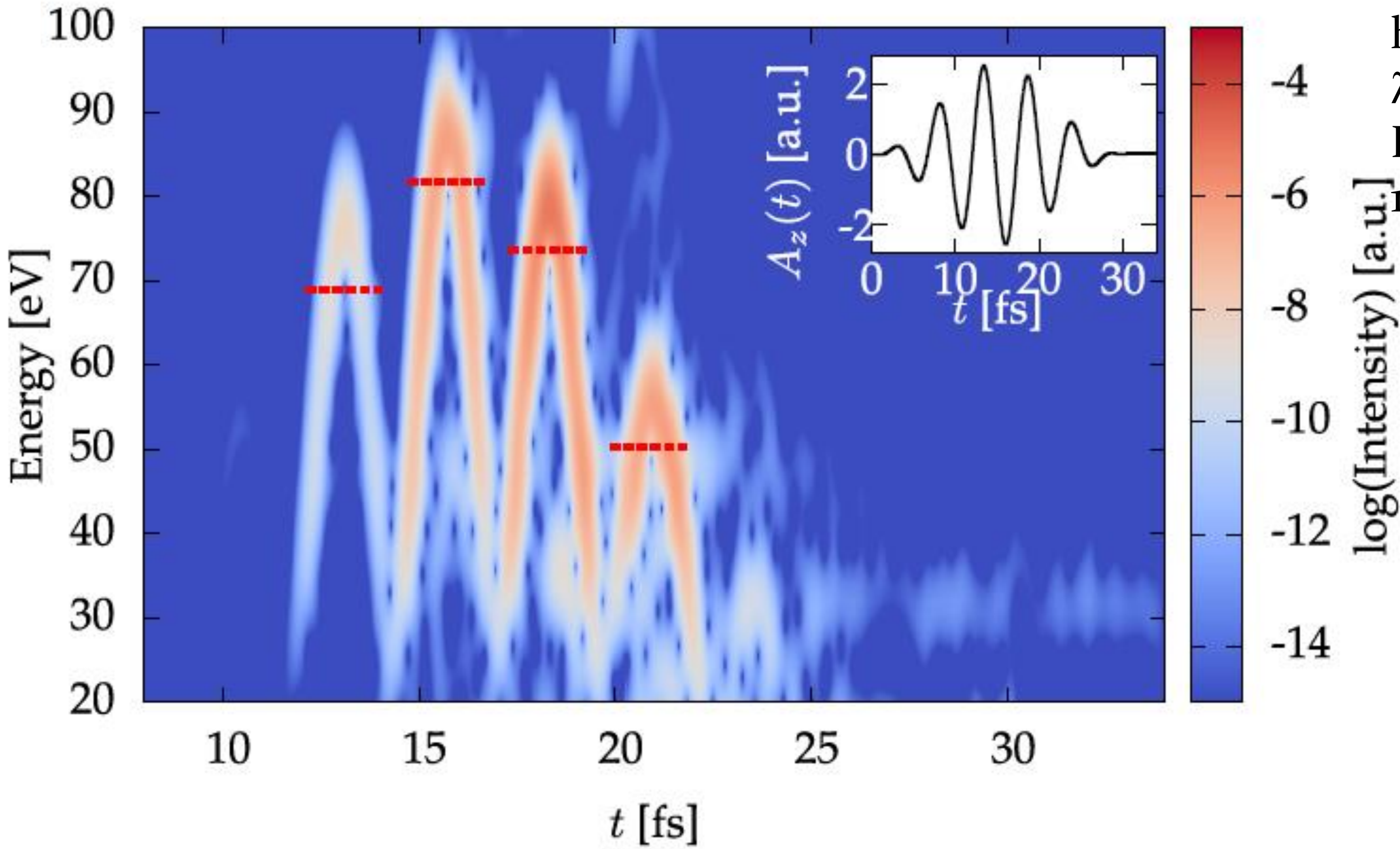
hBN
λ=1600nm
I=1x10^{14} W/cm^2
15fs FWHM

K-point grid
42x42

Energy cutoff from the three-step model

$$E_c = E_w + 3.2\,U_p$$

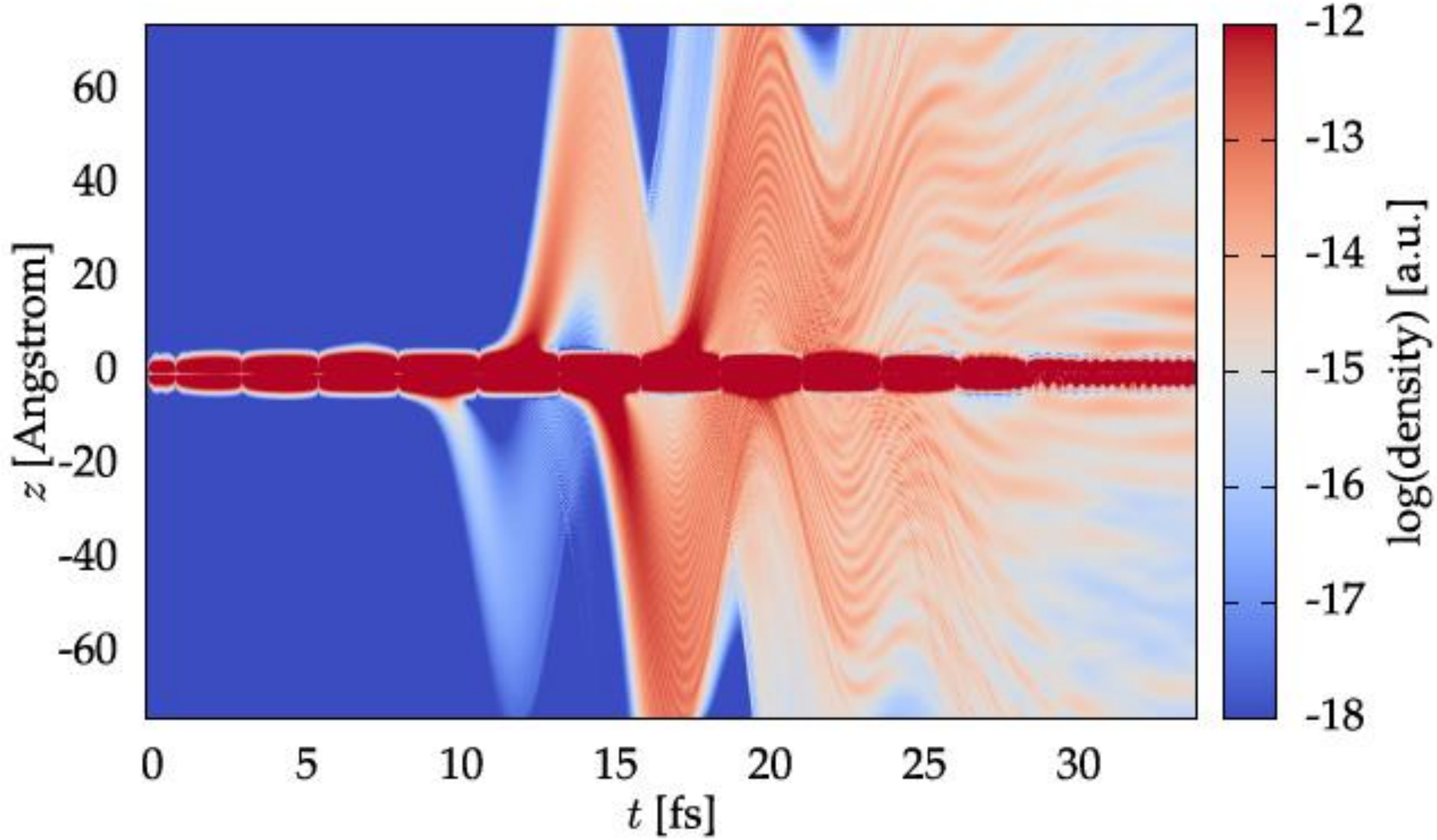
Atomic-like HHG from 2D materials



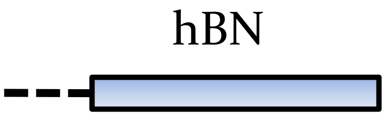
hBN
λ=1600nm
I=1x10^{14} W/cm^2
15fs FWHM

Time-frequency analysis: Well-defined electron trajectories

Atomic-like HHG from 2D materials



hBN
λ=1600nm
I=1x10¹⁴ W/cm²
15fs FWHM

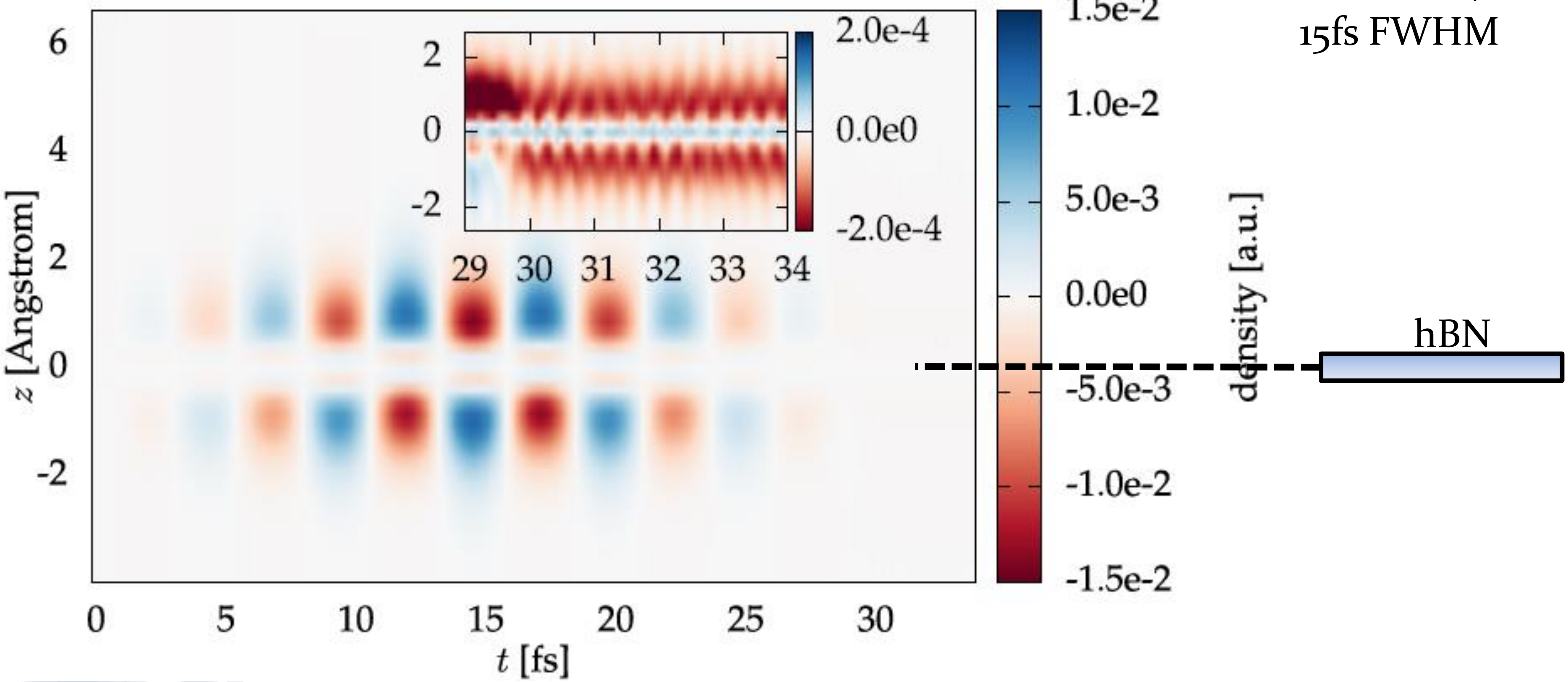


Evolution of the induced electronic density

Beyond IPA: local field effects

mpsd

hBN
λ=1600nm
I=1x10^{14} W/cm^2
15fs FWHM



Evolution of the induced electronic density

Beyond IPA: local field effects

hBN
λ=1600nm
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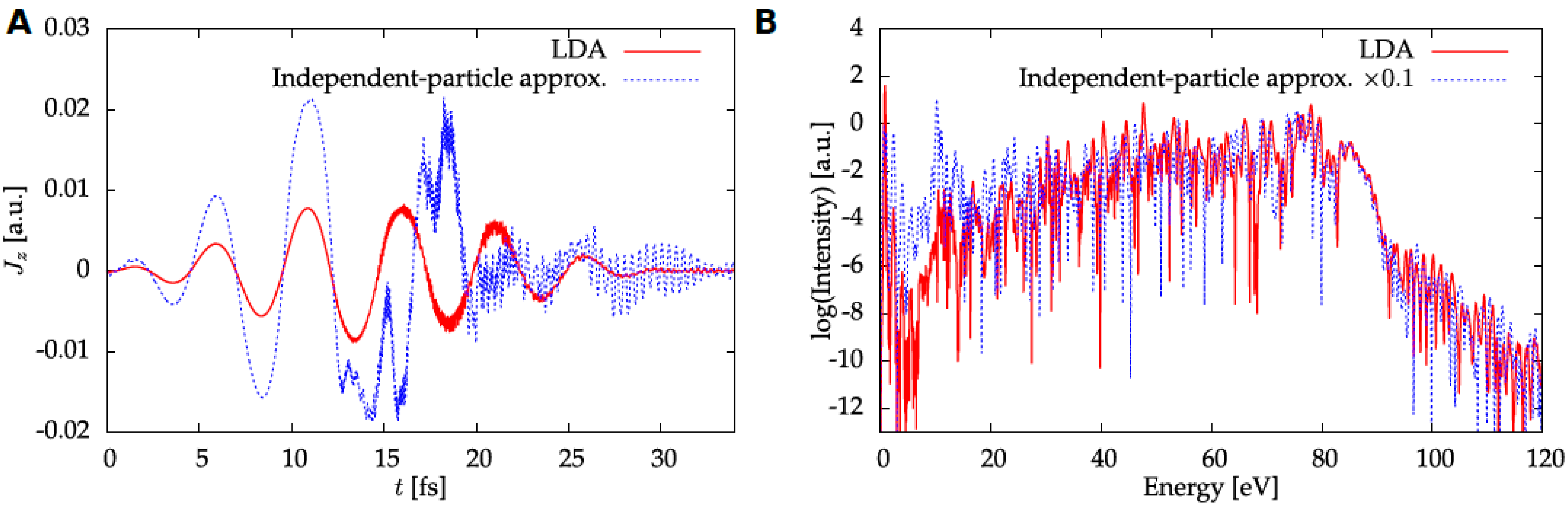

Electronic current

HHG spectra

Wavelength scaling

hBN
I=1x10[14] W/cm[2]
15fs FWHM

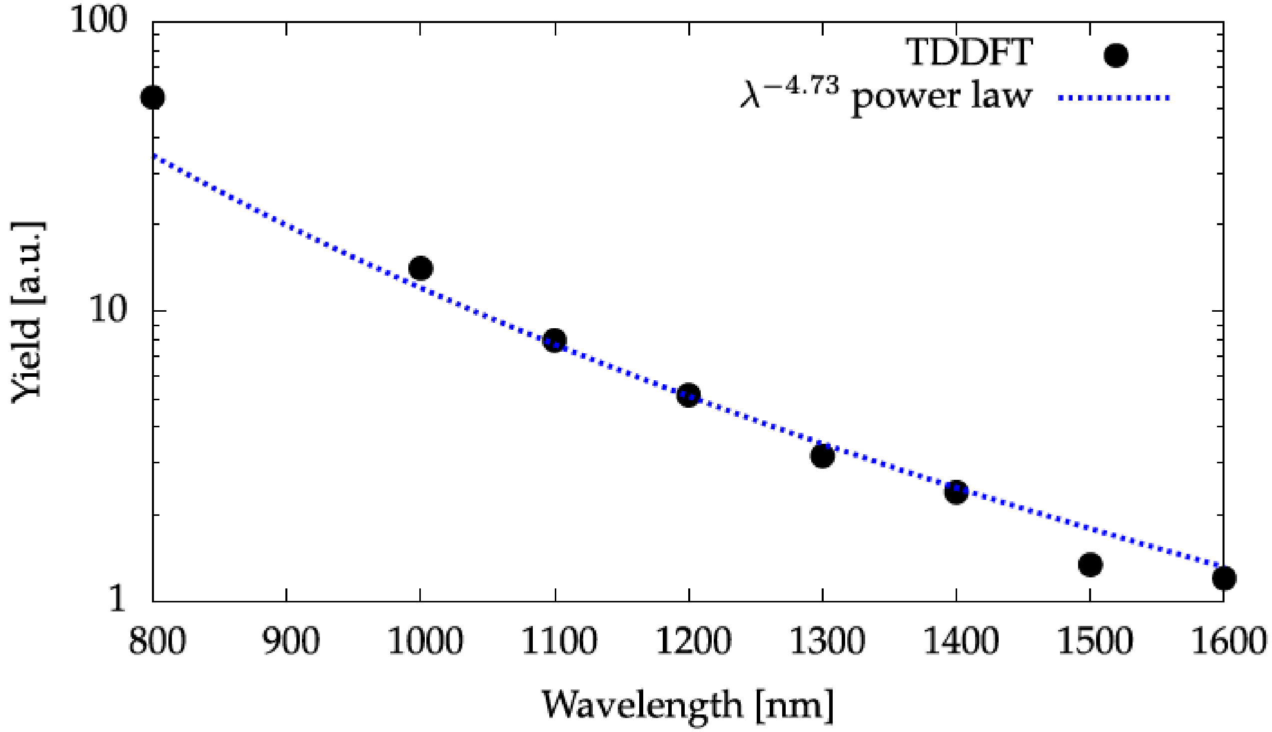

Figure 5: **Calculated wavelength scaling of the harmonic yield.** The harmonic yield obtained from the TDDFT simulations (black point), integrated from 15 eV to 35 eV, as a function of the wavelength, compared to the fitted $\lambda^{-4.73}$ power law.

Conclusions

Possible to suppress interband contribution in favor of HHG yield

Interband and intraband mechanisms react differently to driver ellipticity

Possible to generate circular harmonics in solids, using a single driver field

TDDFT is a powerful predictive tool

[1] N. T.-D. *et al.*, PRL 118, 087403 (2017)
[2] N. T.-D. *et al.*, Nature Comm. 8, 745 (2017)
[3] N. Klemke *et al.*, *Polarization-state-resolved high-harmonic spectroscopy of solids* (Submitted)

Conclusions

Free-standing monolayer materials emit atomic-like HHG for out-of-plane driving fields

First and third steps of the three-step model are modified:

- Local fields play an important role

- Wavelength-scaling is better than in atoms

2D materials offer a platform to study both atomic and bulk HHG

N. T.-D. and A.R. *Atomic-like high-harmonic generation from two-dimensional materials.* Sci. Adv. 4, eaao5207 (2018).

Thank you for your attention