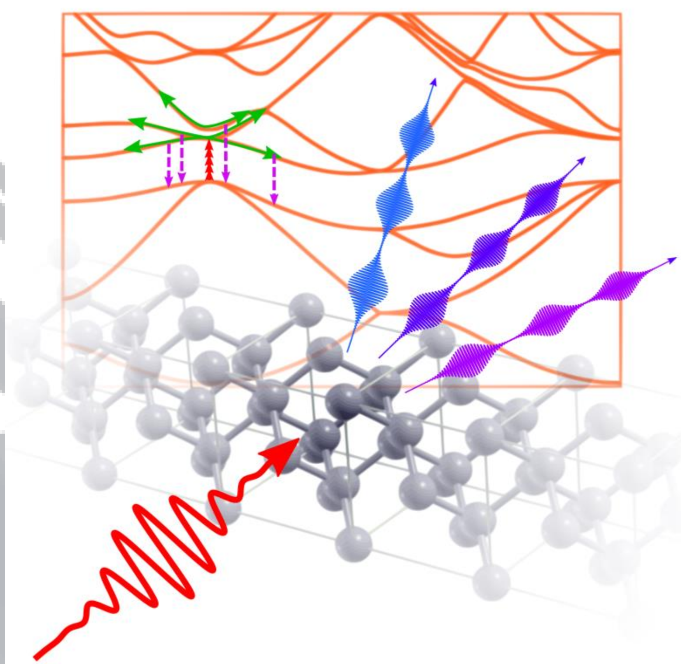
High-Harmonic Generation Spectra of Solids

Nicolas Tancogne-Dejean

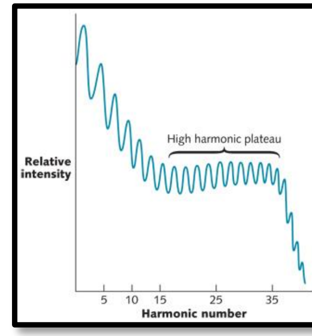


Collaborators: O. D. Mücke, F. X. Kärtner, Angel Rubio

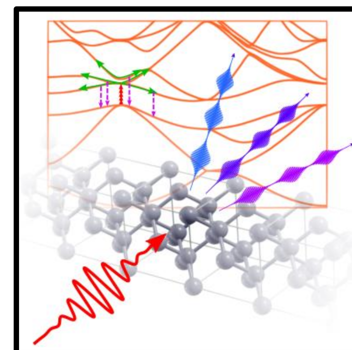
Max Planck Institute for the Structure and Dynamics of Matter

Outline

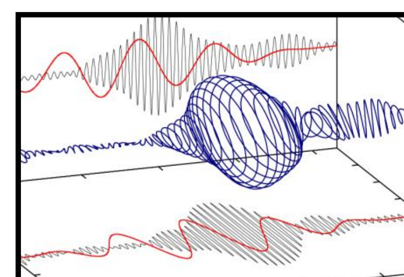
- High-harmonic generation (HHG)



- Impact of the band-structure



- Ellipticity dependence



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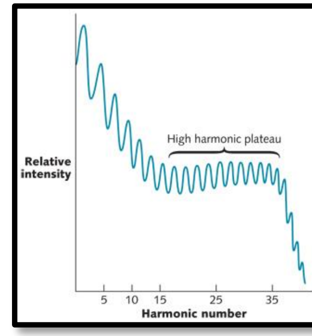

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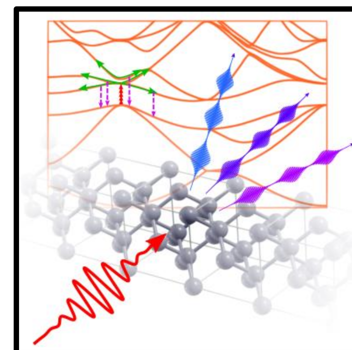

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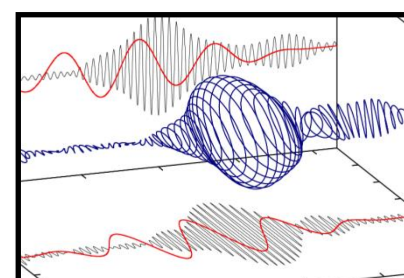
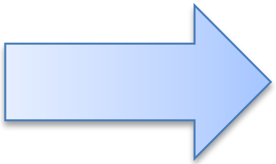


Response to a perturbation

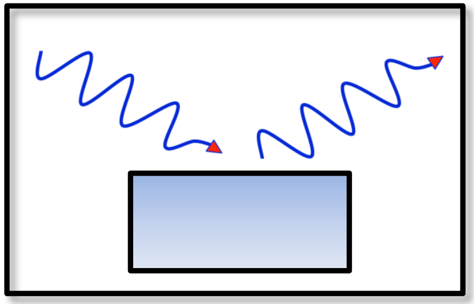


Perturbation

Electric field

Response

Polarisation



Linear Response | **Nonlinear Response**

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

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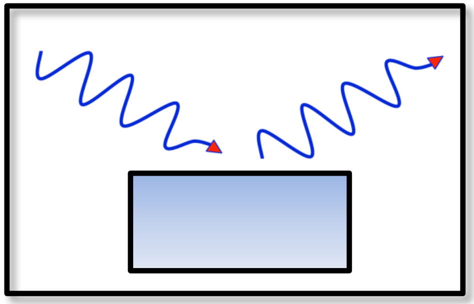

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For weak lasers

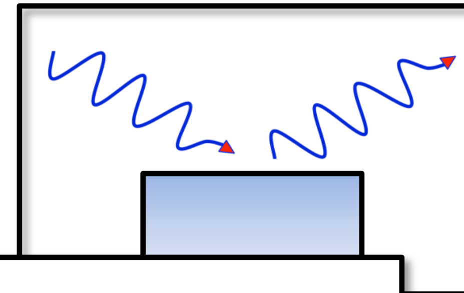
($< 10^{11}$ W/cm^2)

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

Perturbative regime

Response to a perturbation

Perturbation	Response
Electric field	Polarisation

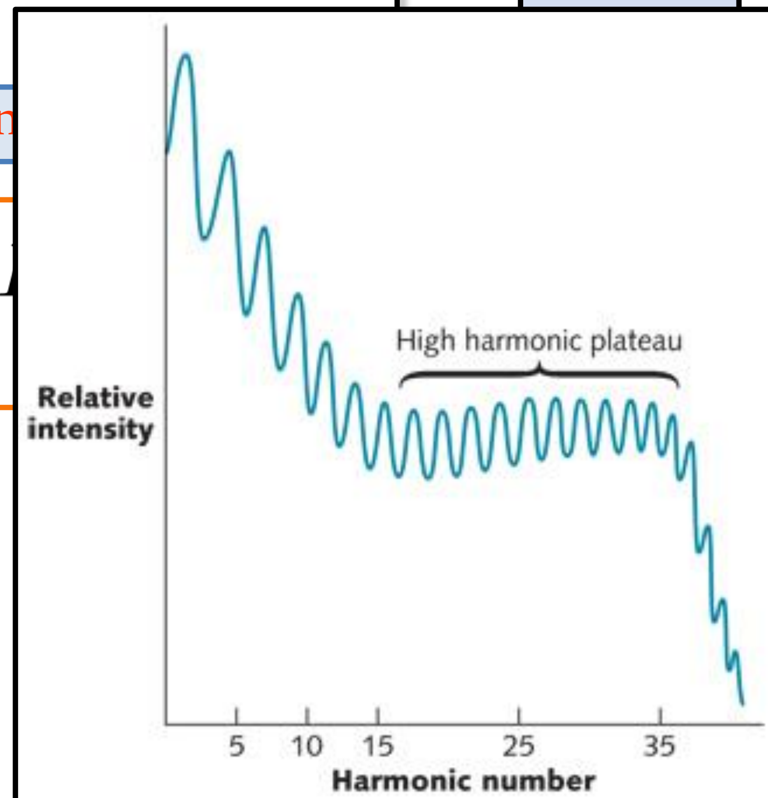


Linear Response

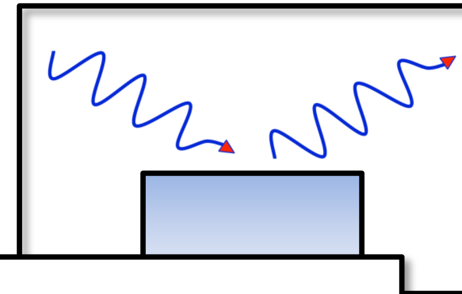
Non

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

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



Response to a perturbation



Perturbation

Electric field

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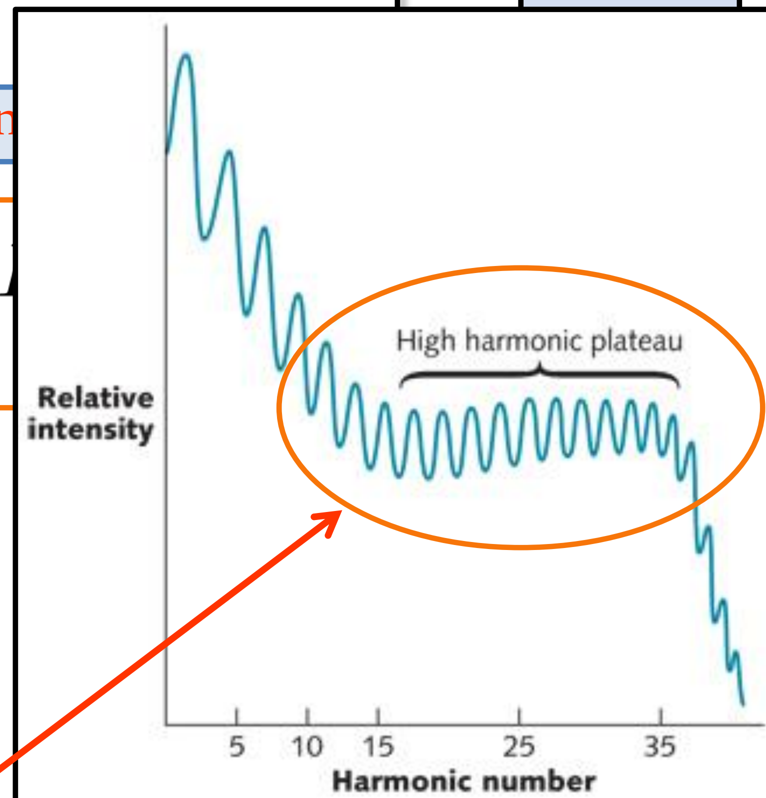
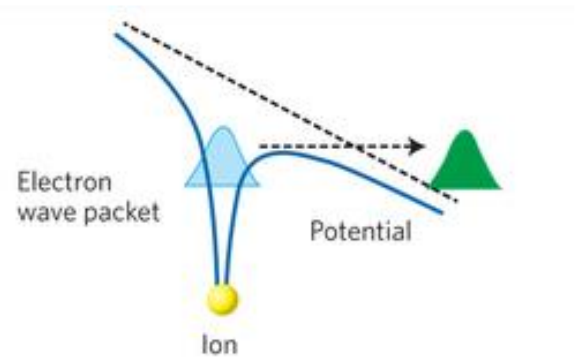


Non-perturbative regime

HHG in atoms: three-step model

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



1. Tunneling

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

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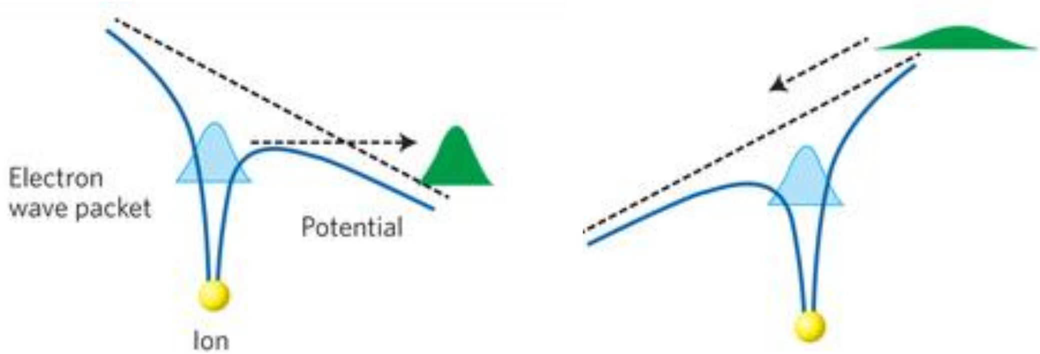

1. Tunneling 2. Acceleration by the field

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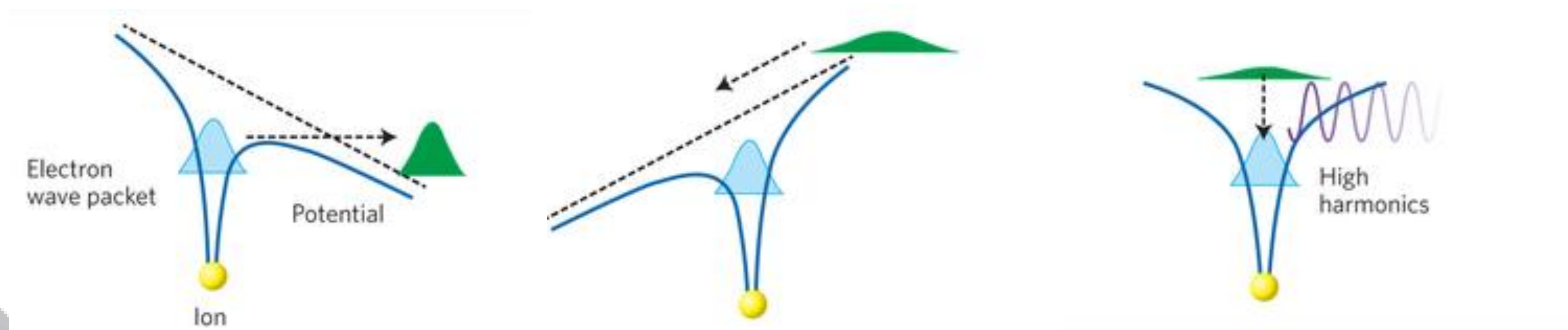


1. Tunneling 2. Acceleration by the field 3. Recombination

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

And 30 years later… HHG in solids

LETTERS

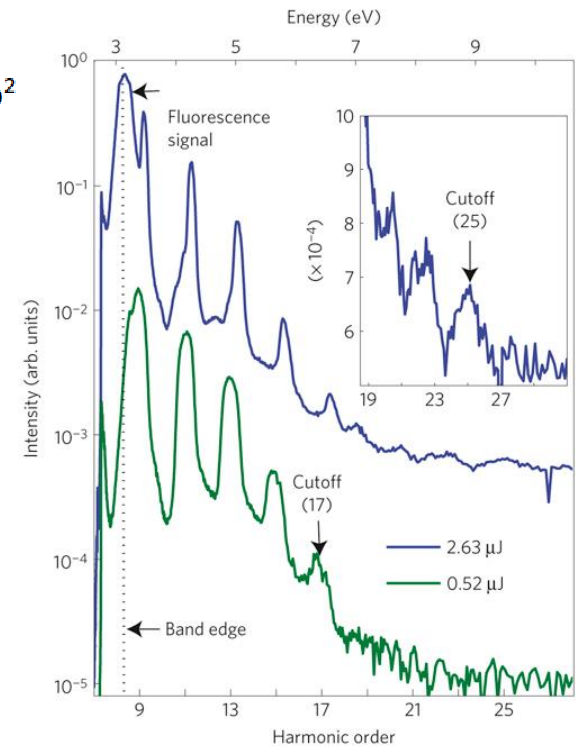
PUBLISHED ONLINE: 5 DECEMBER 2010 | DOI: 10.1038/NPHYS1847

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



High-harmonic generation (HHG) in solids

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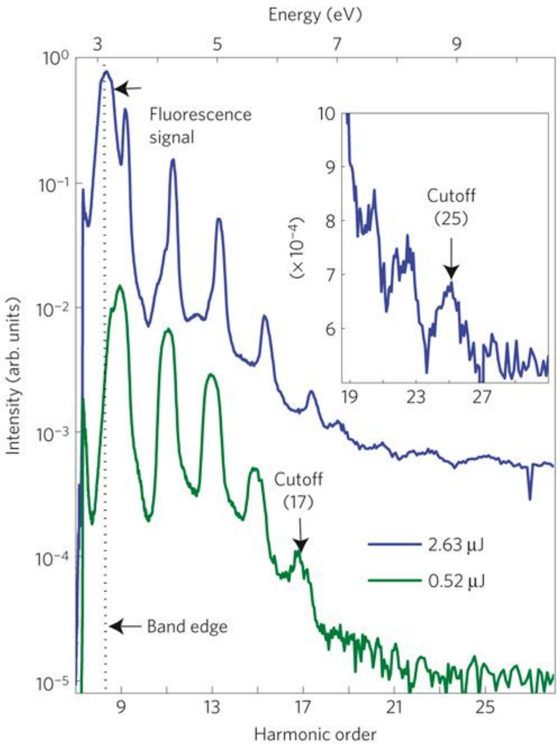


Marangos, Nat. Phys. 7, 97 (2011)
Schubert *et al.*, Nat. Phot. 8, 119 (2014).
Kim et al., Nat. Phot. 8, 92 (2014).
Hohenleutner *et al.*, Nature 523, 572 (2015).
Vampa *et al.*, Nature 522, 462 (2015).
Luu *et al.*, Nature 521, 498 (2015).
Vampa *et al.*, PRL 115, 193603 (2015).

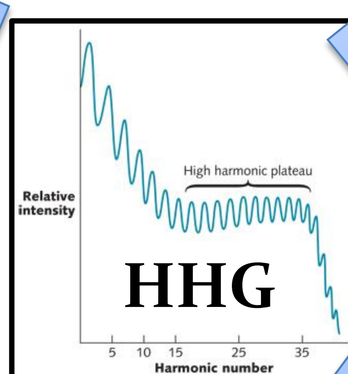
Some applications of HHG in solids

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

Table-top synchrotron

Electron-hole recollisions in real time

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



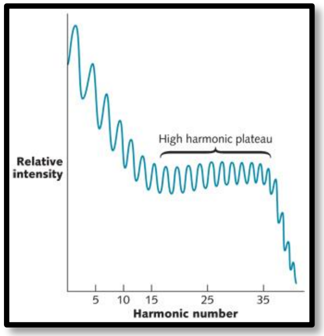
All-optical band-structure reconstruction

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

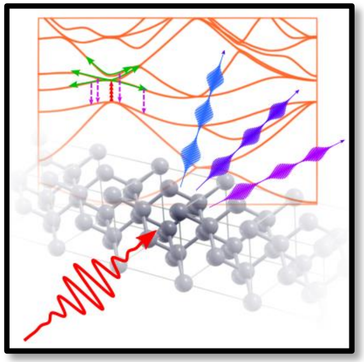
Quantum-logic at optical clock-rates

Outline

- High-harmonic generation (HHG)



- HHG in solids: Impact of the band structure



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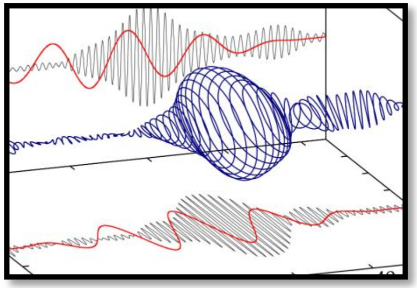

Understanding HHG in solids

What is the microscopic mechanism responsible for HHG in solids?

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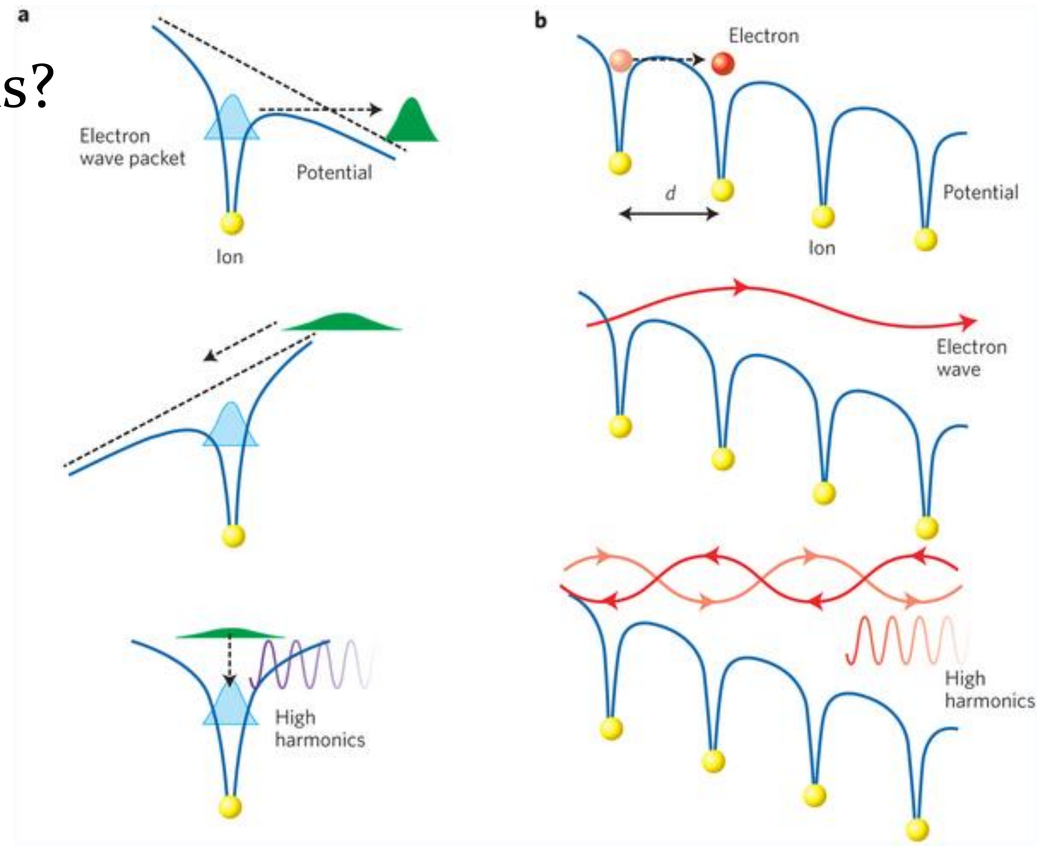
A similar mechanism as in atoms?



From Kim *et al.* Nature Photonics **8**, 92 (2014)

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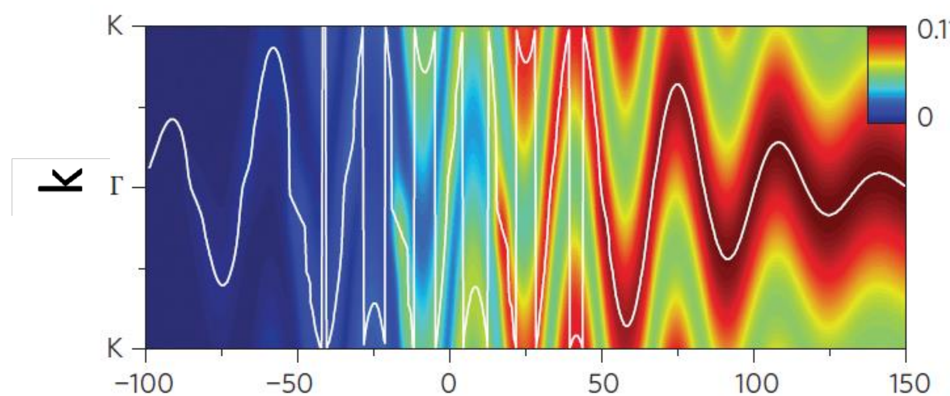
Dynamical Bloch oscillations?



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Understanding HHG in solids

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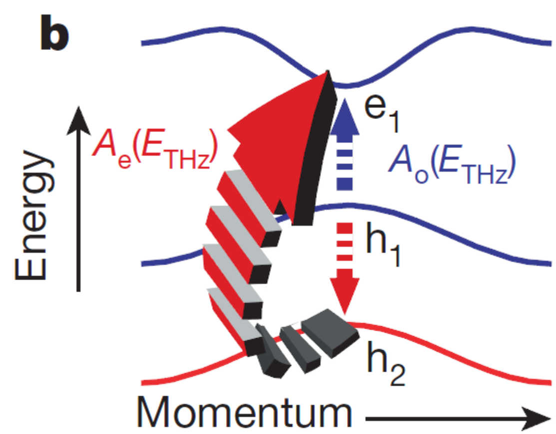
Interband transitions?



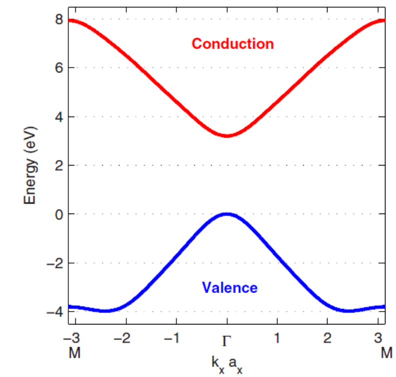
From Hohenleutner *et al.* Nature **523**, 572 (2015)

How many bands are contributing?

Understanding HHG in solids

What is the microscopic mechanism responsible for HHG in solids?

Two-band model?



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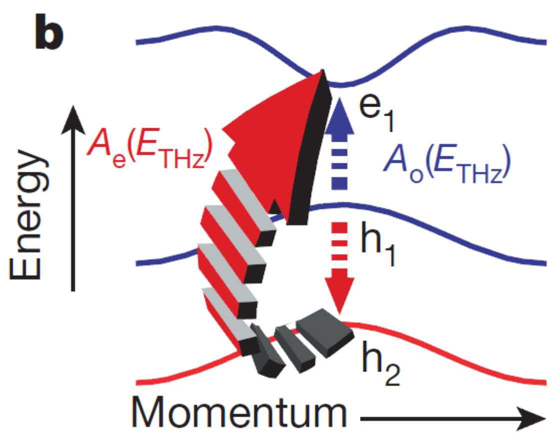
Interband transitions?



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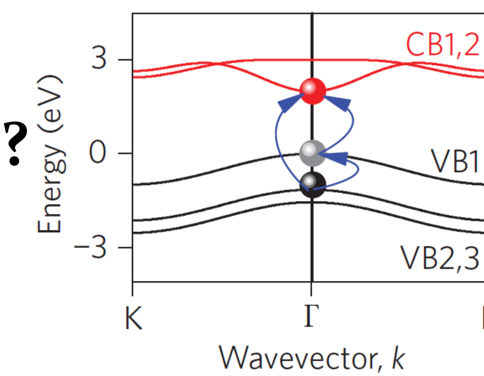
Five-band model?



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Nature Photonics **8**, 119 (2014)

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

mpsd

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

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

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

[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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we obtain

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

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

[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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- No HHG from an homogeneous electron gas (parabolic bands)

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Some exact analytical results

mpsd

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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

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

Independent-particle approximation:

$$i\frac{\partial}{\partial t}\phi_i(\mathbf{r},t) = \left(-\frac{\nabla^2}{2} + v_{\mathrm{ext}}(\mathbf{r},t) + v_{\mathrm{H}}[n](\mathbf{r},\underline{t_0}) + v_{\mathrm{xc}}[n](\mathbf{r},\underline{t_0})\right)\phi_i(\mathbf{r},t)$$

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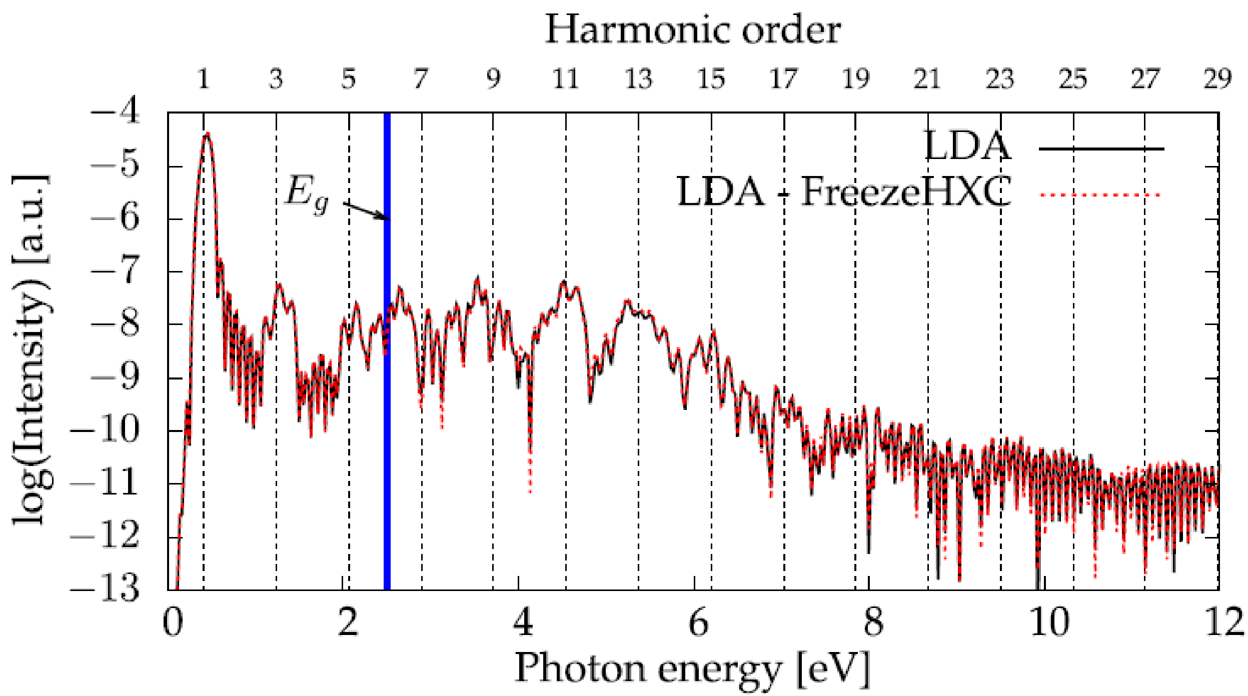
Correlation effects in HHG

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=10¹¹ W/cm²

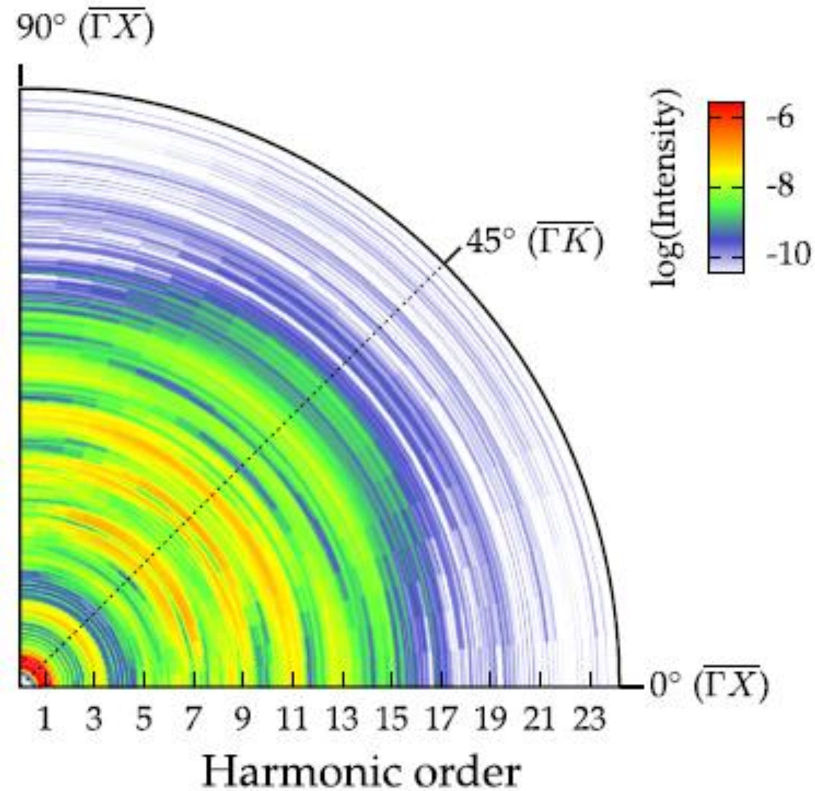
[1] N. T-D *et al.*,PRL 118, 087403 (2017)

Anisotropy of the HHG in solids

Electrons only explore a restricted portion of the Brillouin zone

-> HHG emission is anisotropic, even in cubic materials

Calculated TDDFT anisotropy map of the HHG spectra obtained by rotating the laser polarization around the [001] crystallographic direction



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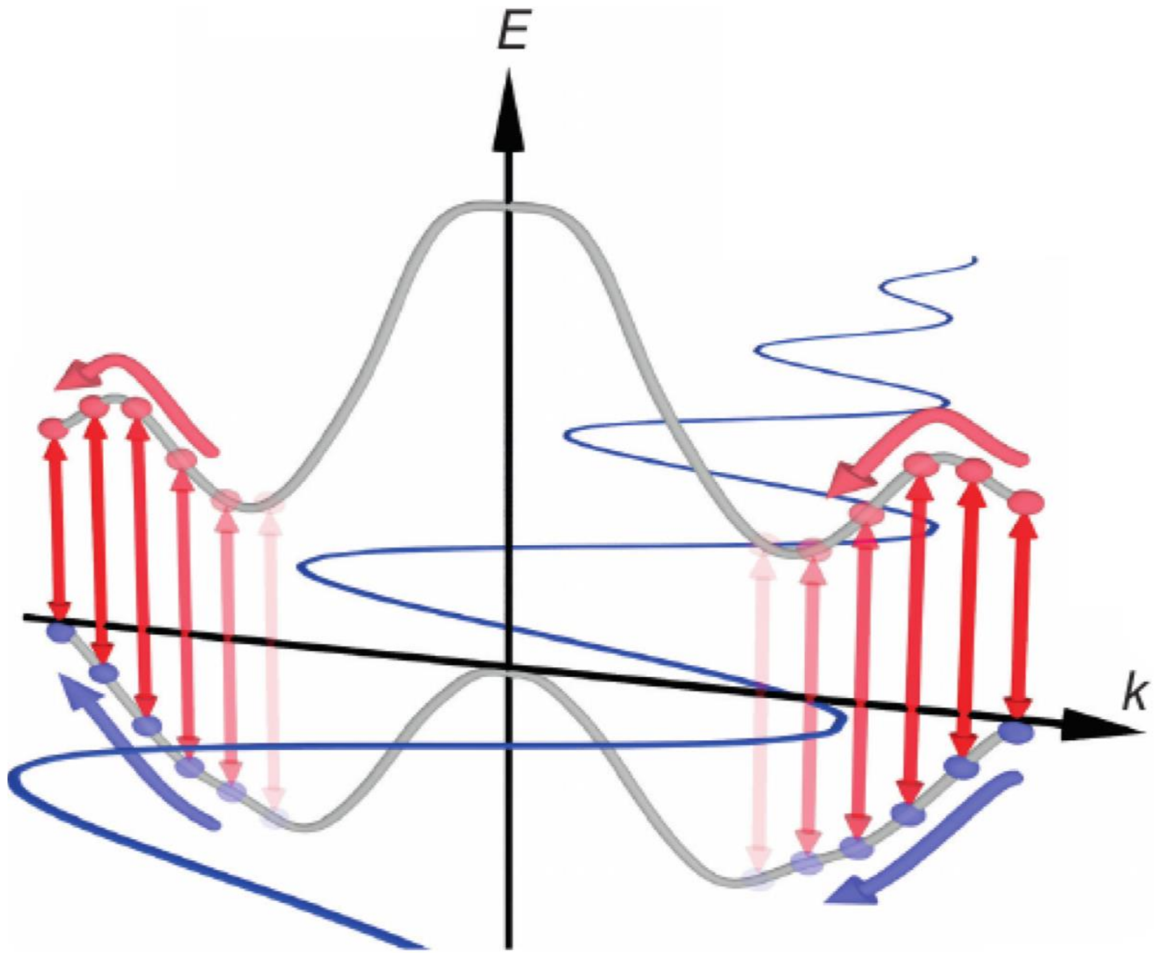
Interbandvs Intraband mechanism

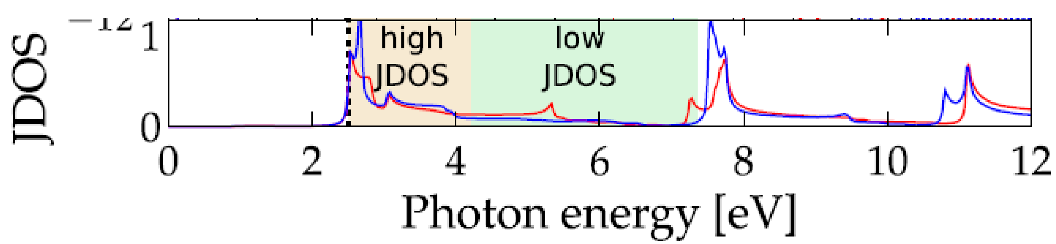


Adapted from Langer et al., Nature 533, 225 (2016)

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)



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

Interband *vs* Intraband mechanism

mpsd

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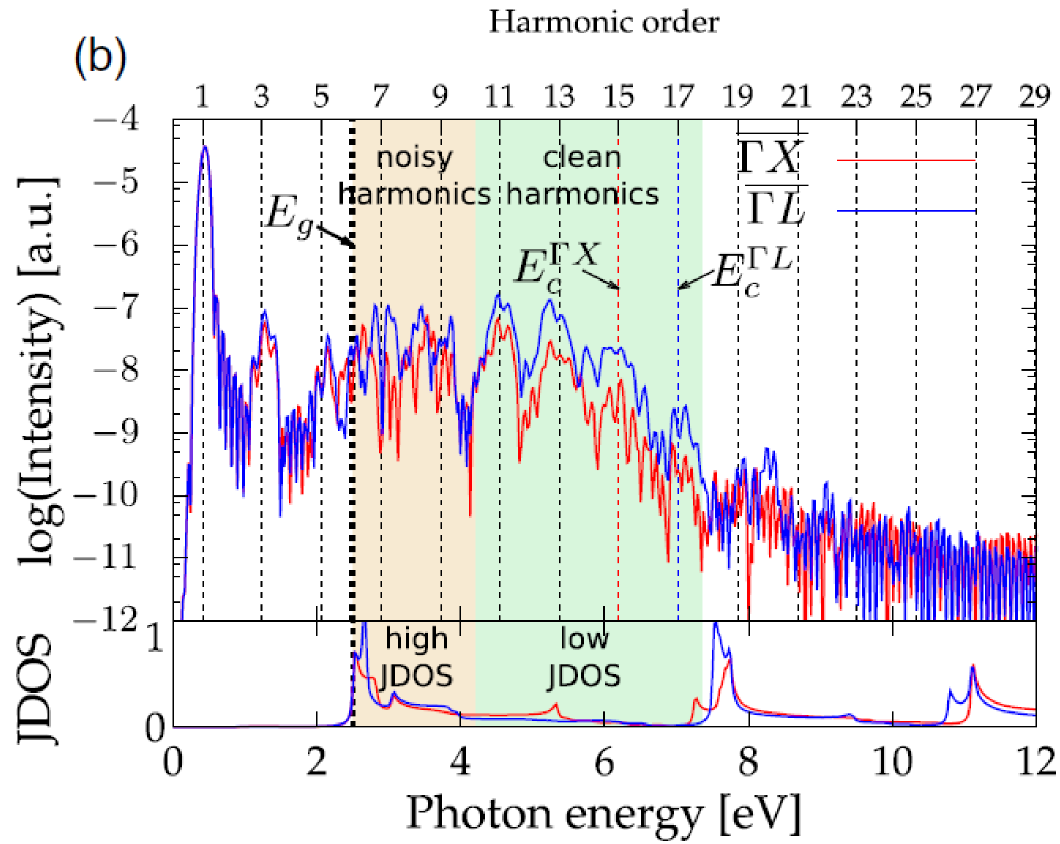
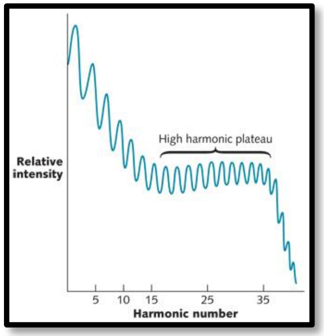


- Low JDOS: interband contribution is suppressed
- HHG yield improved when interband is suppressed
- Toward band-structure engineering to improve HHG in solids

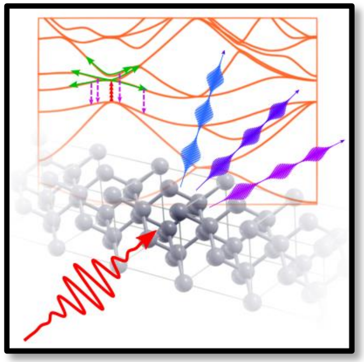
[1] N. T-D *et al.*,PRL 118, 087403 (2017)

Outline

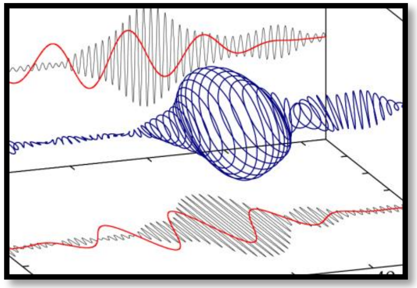
- High-harmonic generation (HHG)



- Impact of the band-structure

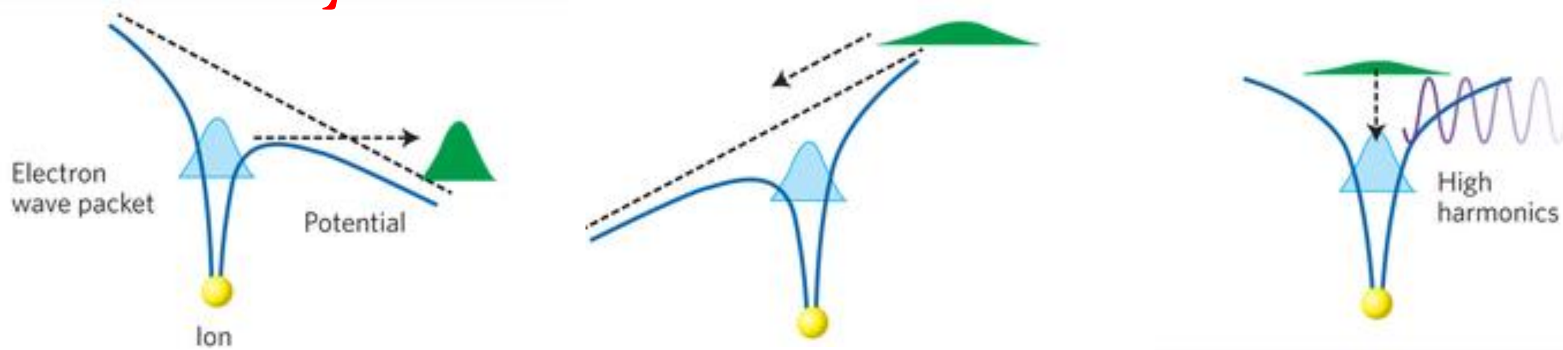


- **Ellipticity dependence**

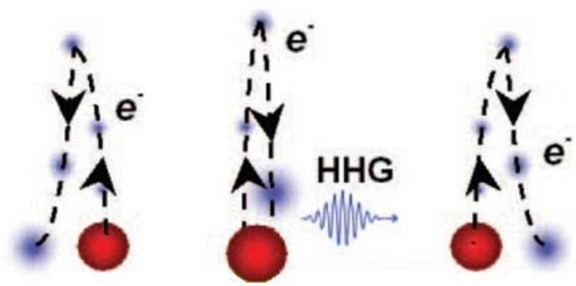


Ellipticity dependence in gases

In atomic gases, circular light suppresses the harmonic yield



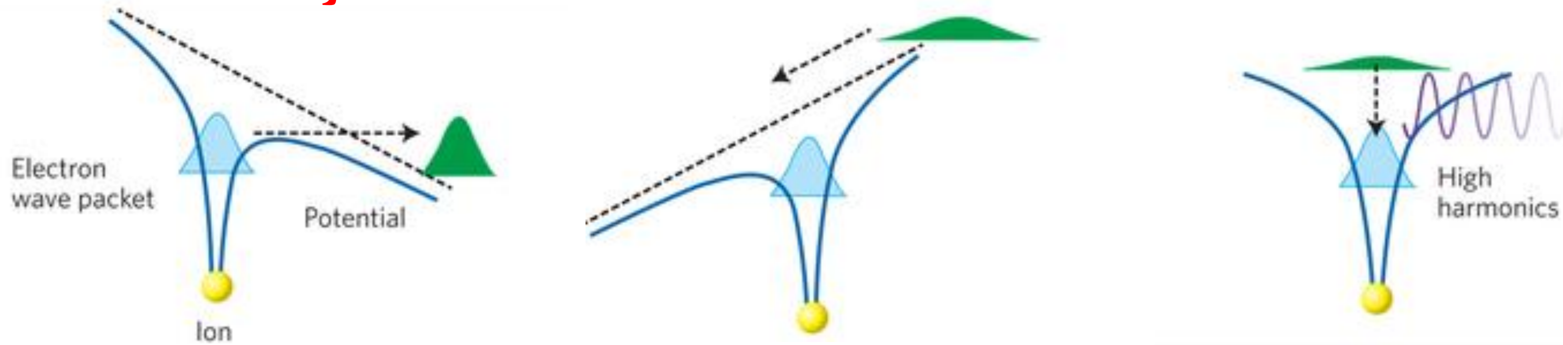
Electrons acquire a transversal momentum and "misses" the parent ion.
No recombination, no harmonic emission



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

Ellipticity dependence in gases

In atomic gases, circular light suppresses the harmonic yield



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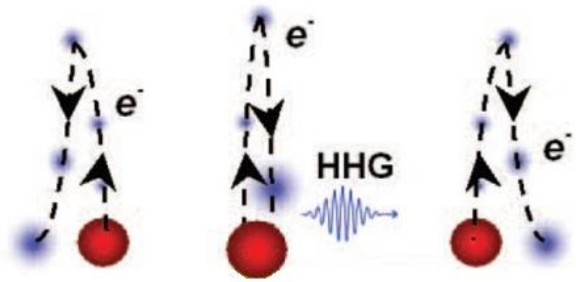
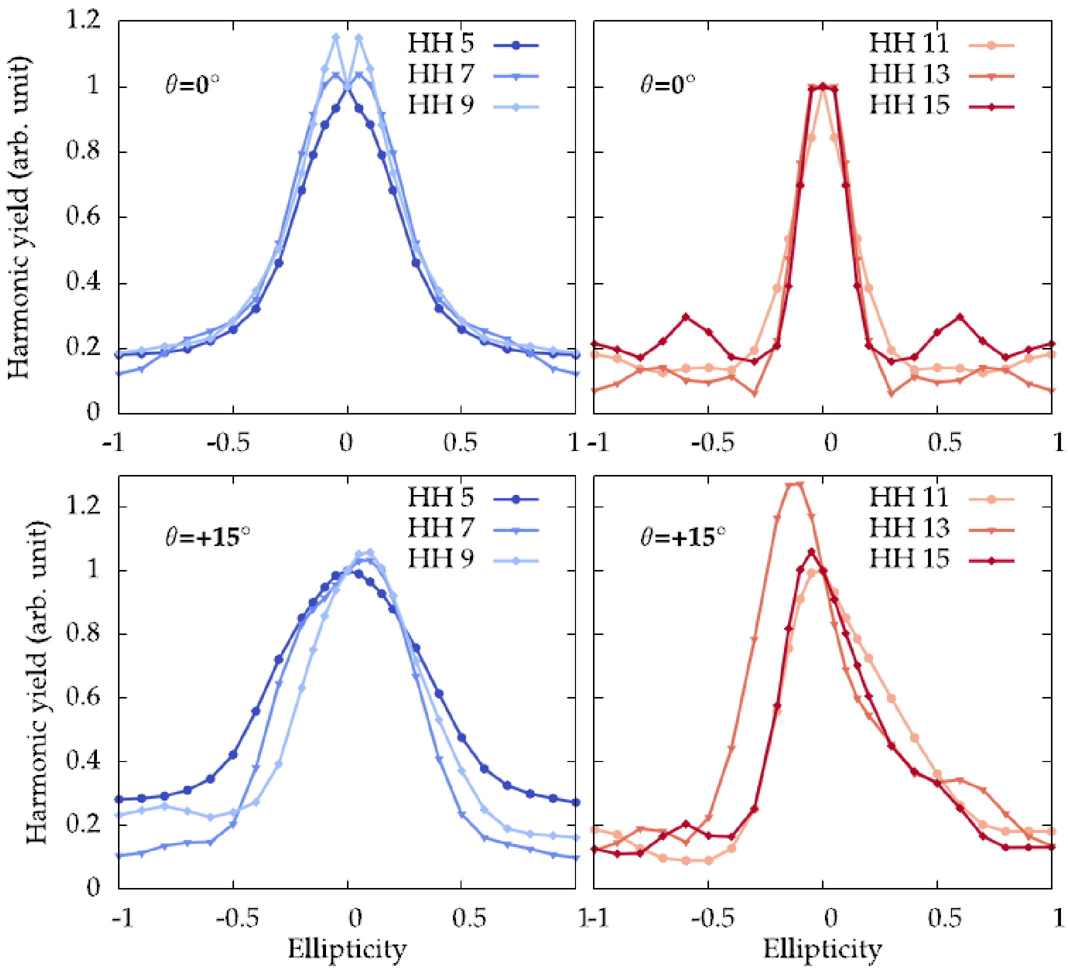


B. Shan *et al.*, J. Mod. Opt. **52**, 277 (2005)

Not the case in solids !

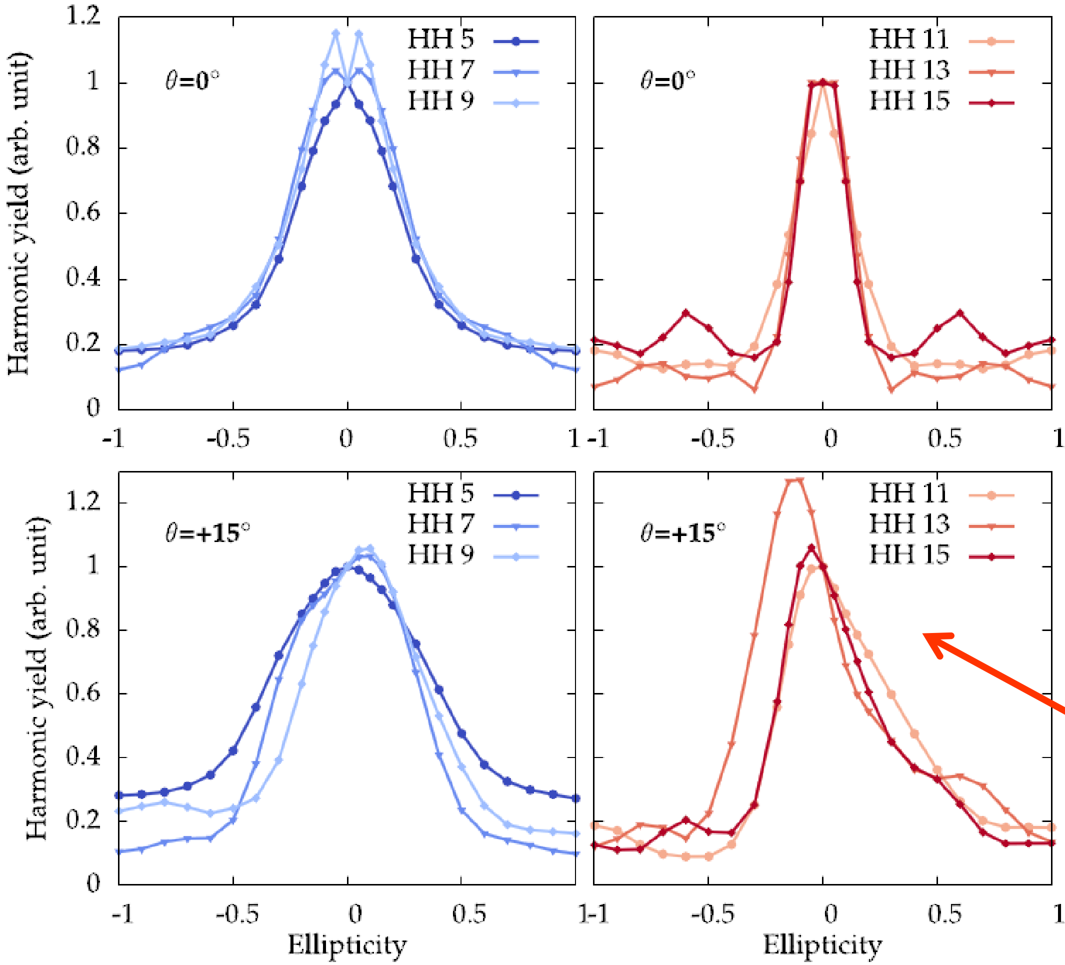
Ellipticity dependence of HHG in solids



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

[1] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

Ellipticity dependence of HHG in solids

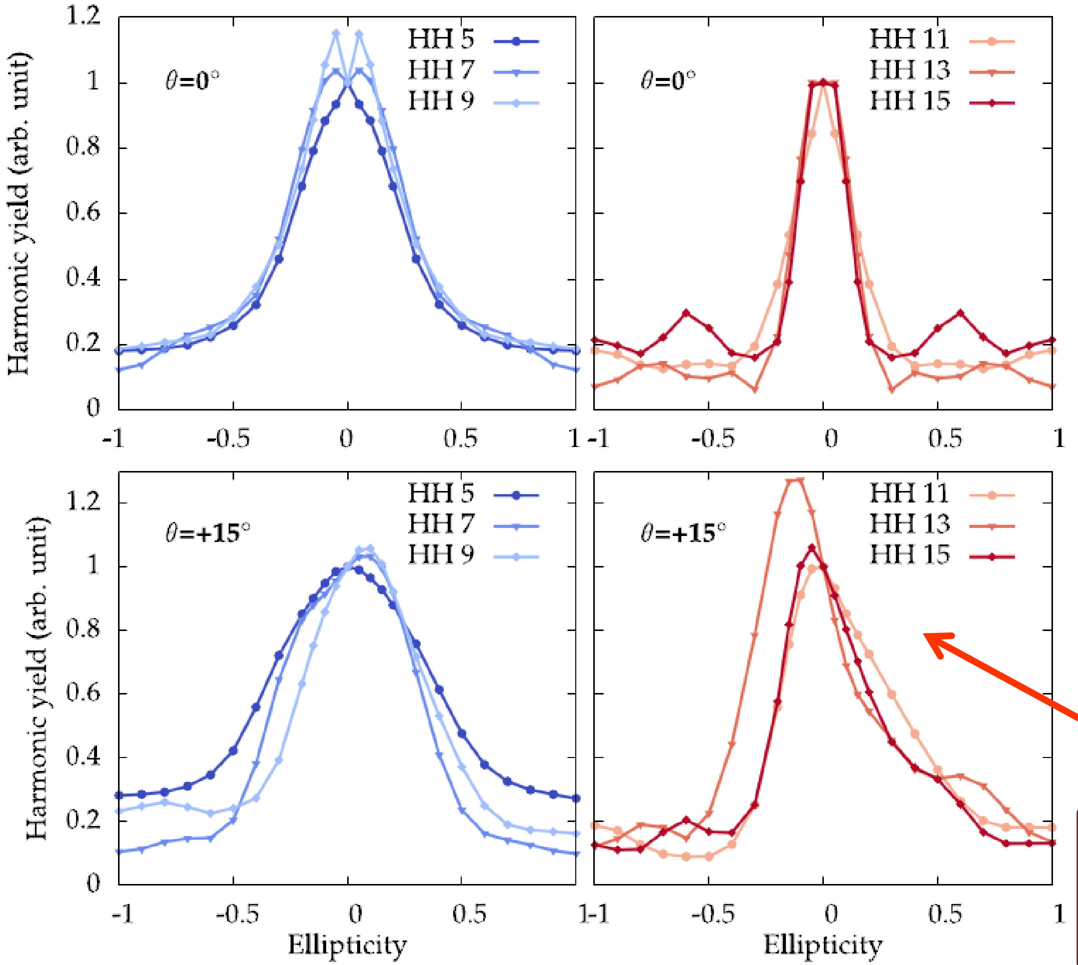


Bulk Si
λ=3000nm
$I=3x10^{12}\ W/cm^2$
25fs FWHM

Anisotropic ellipticity profiles

[1] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

Ellipticity dependence of HHG in solids

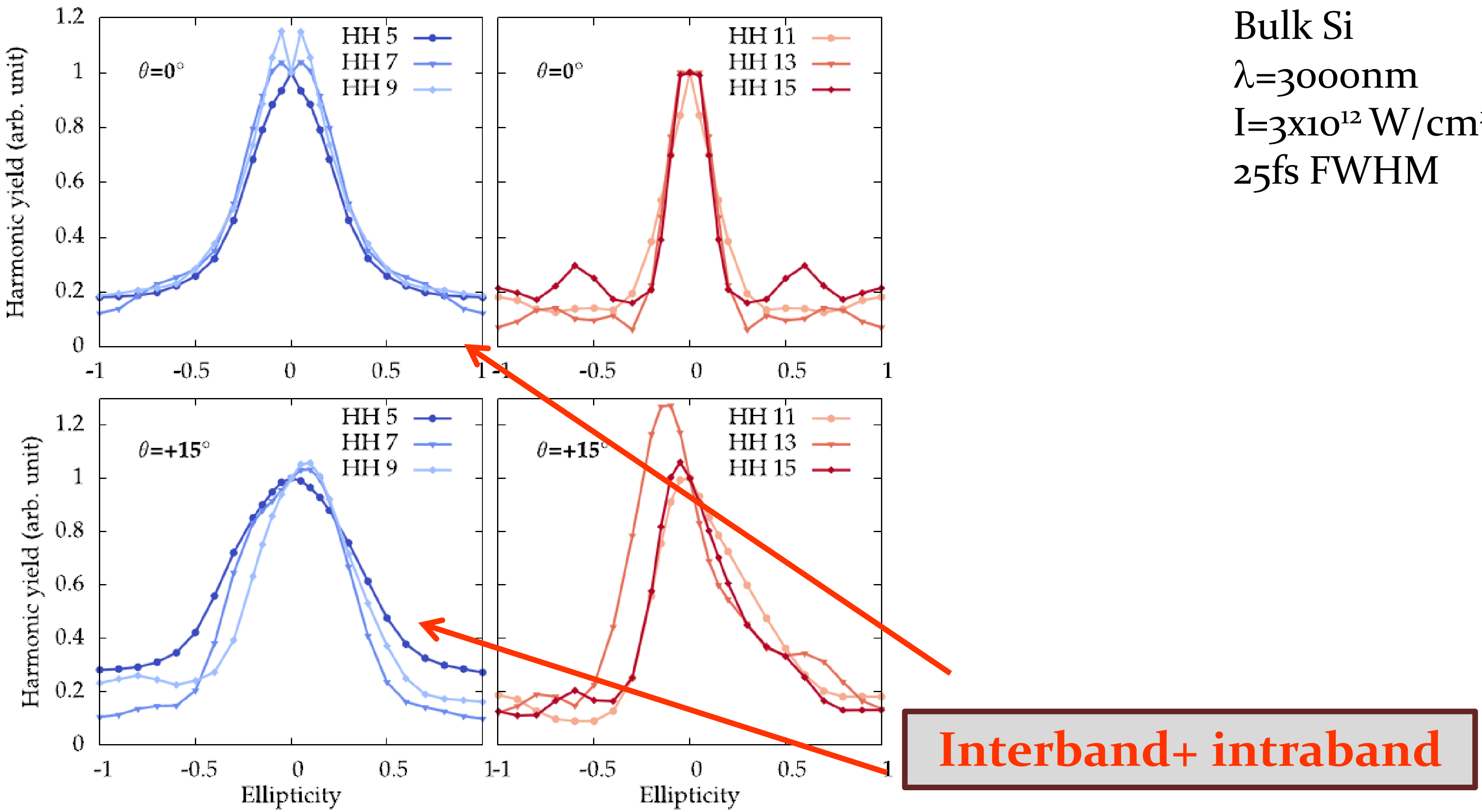


Bulk Si
λ=3000nm
I=3x10¹² W/cm²
25fs FWHM

Mostly from intraband

[1] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

Ellipticity dependence of HHG in solids



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

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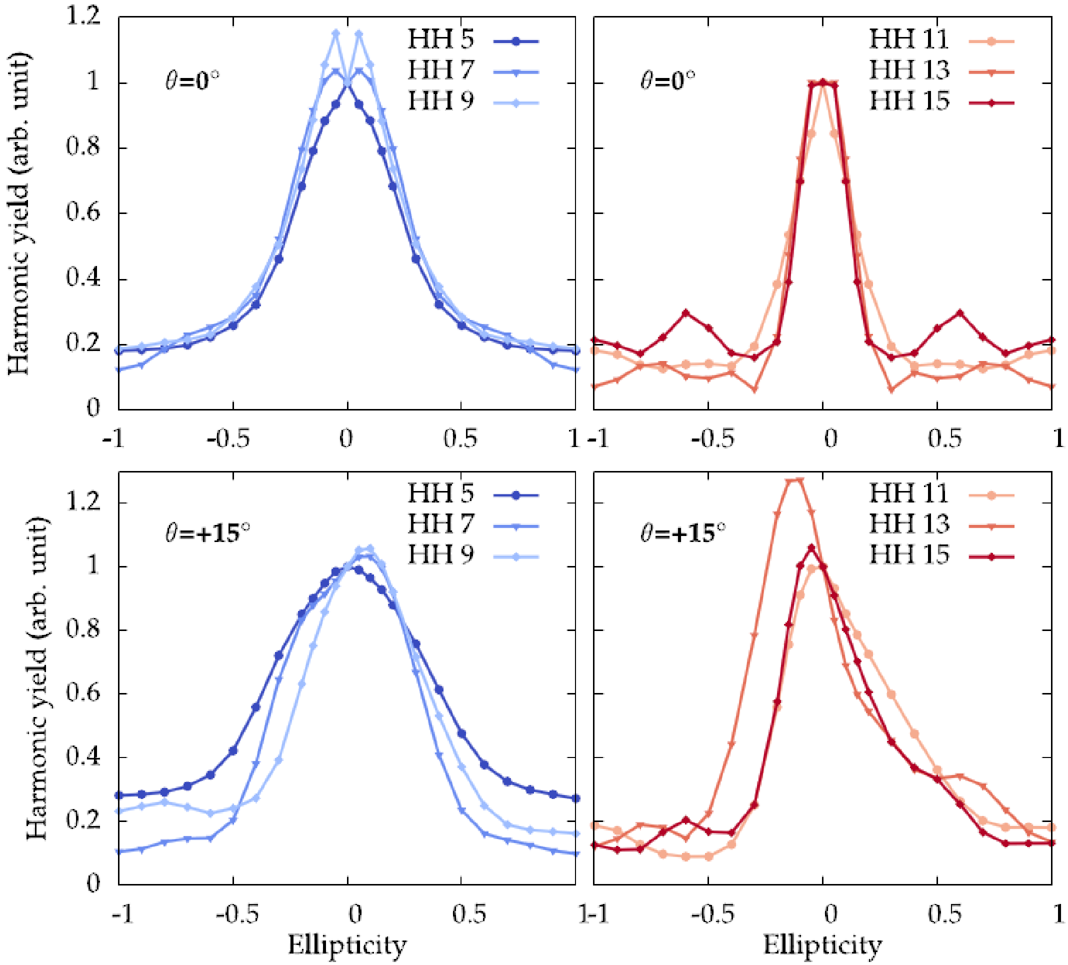
Ellipticity dependence of HHG in solids

mpsd

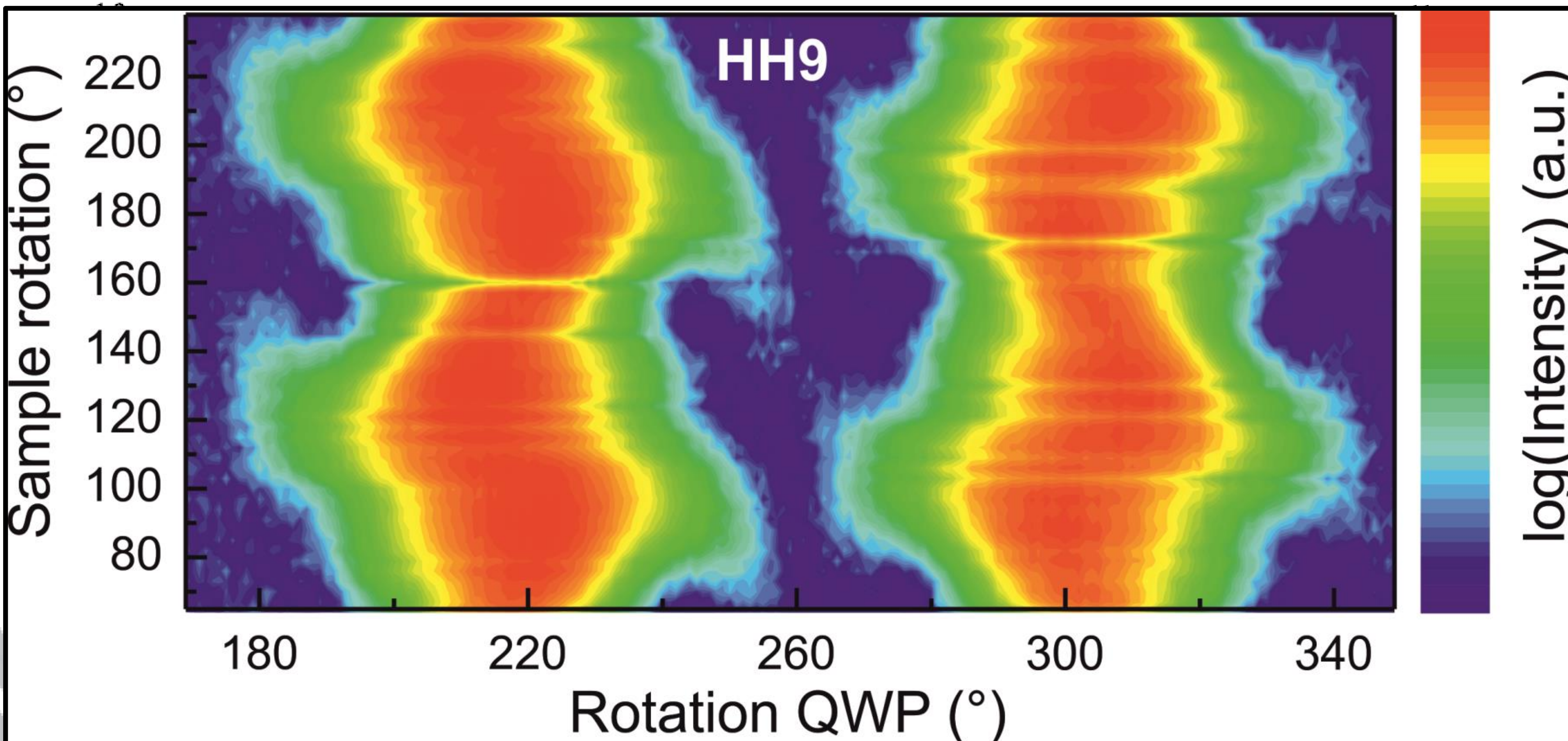


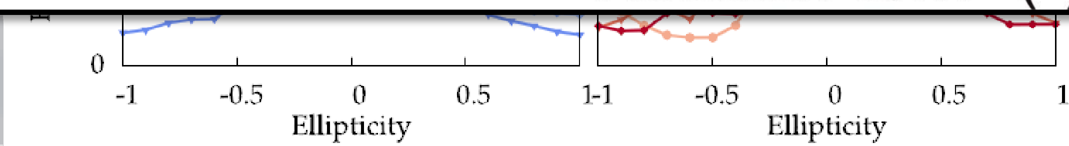
Bulk Si
λ=3000nm
I=3x10[12] W/cm²
25fs FWHM

Interband and intraband react differently the driver ellipticity

[1] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

Ellipticity dependence of HHG in solids



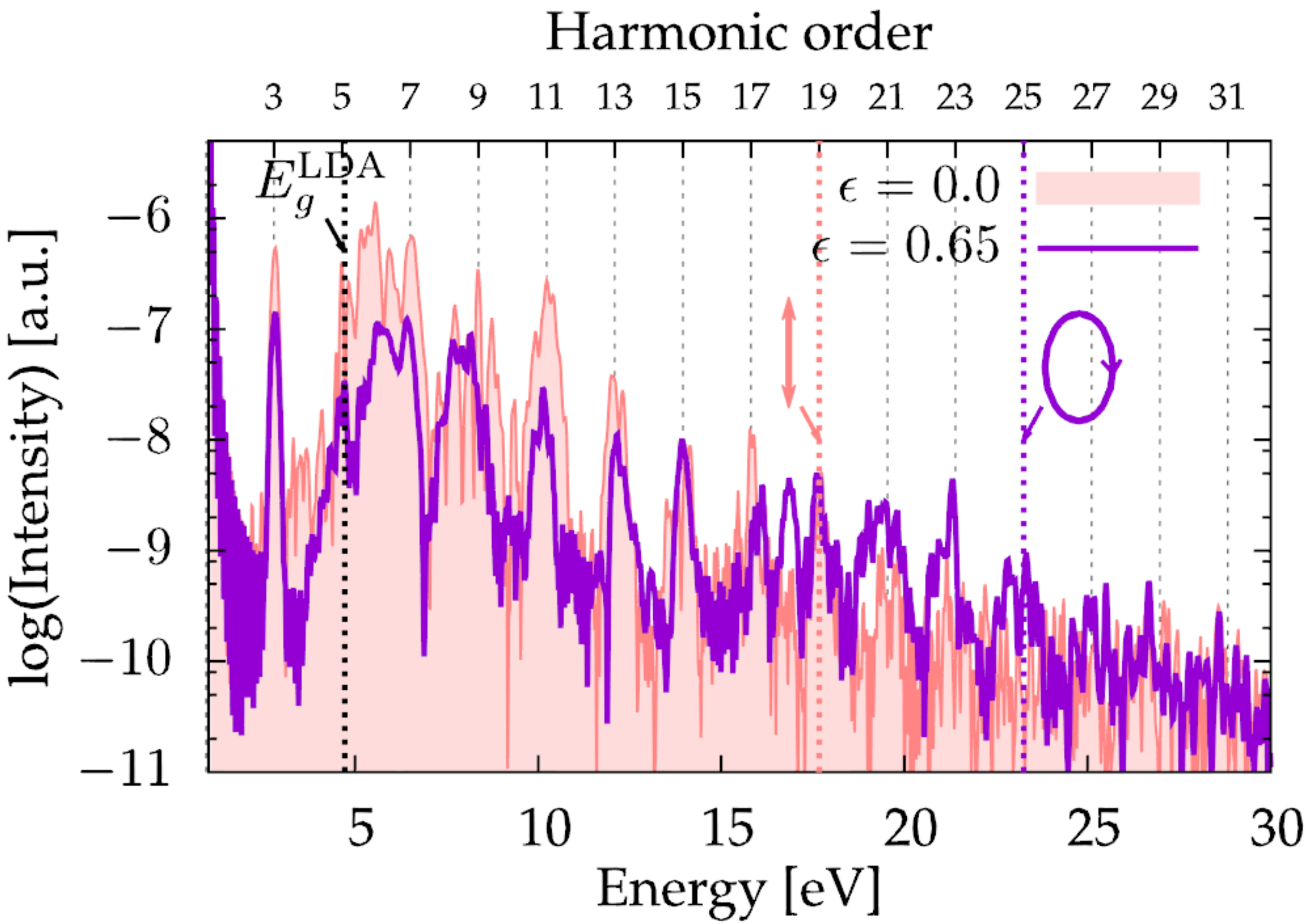


ellipticity

[1] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

Energy cutoff increase from ellipticity

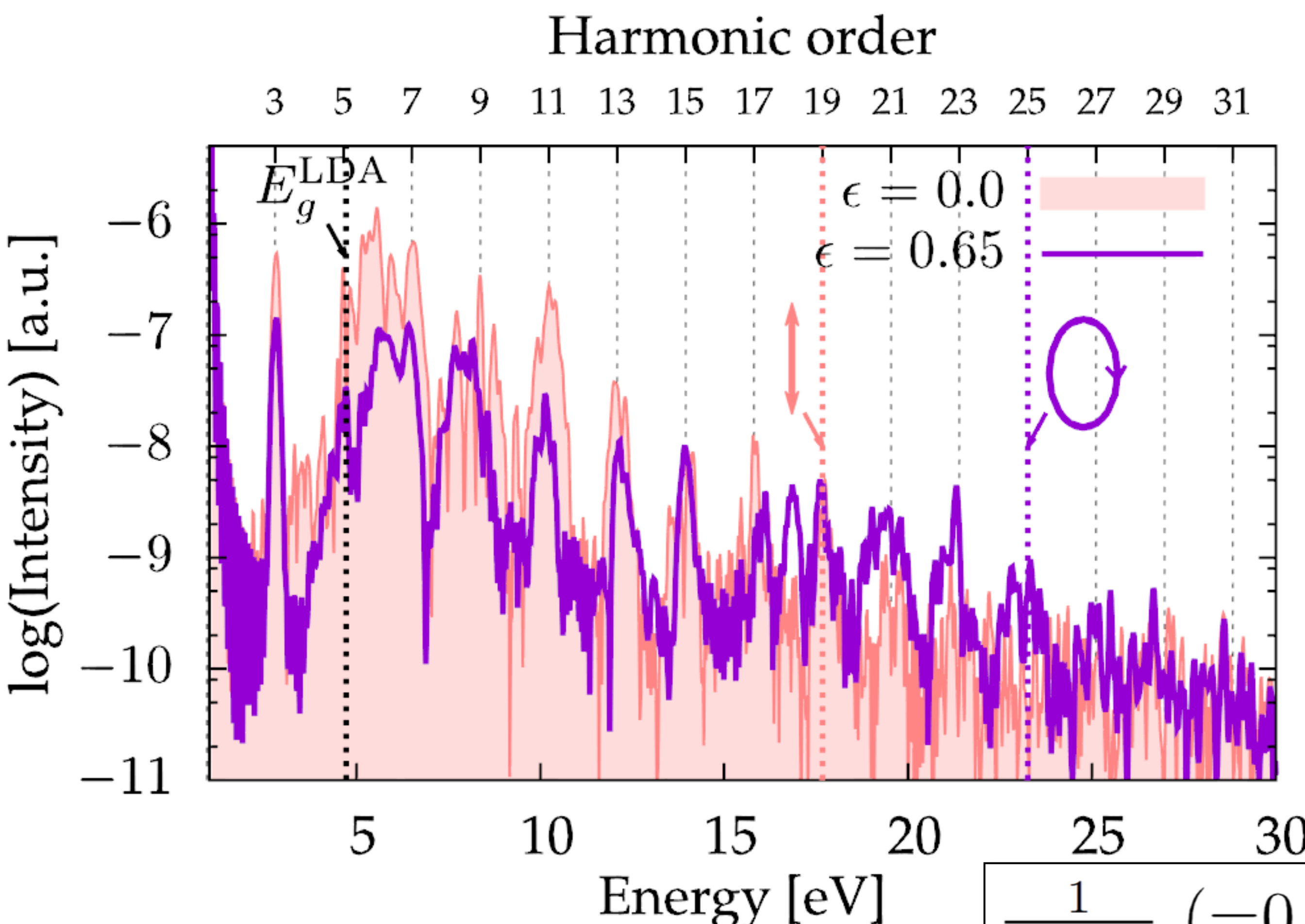
mpsd



Bulk MgO
λ=1333nm
I=3x10[12] W/cm[2]
50fs FWHM

[1] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

Energy cutoff increase from ellipticity



Bulk MgO
λ=1333nm
I=3x10[12] W/cm[2]
50fs FWHM

$$\frac{1}{\sqrt{1+\epsilon^2}} \; (=0.84 \text{ for } \epsilon = 0.65)$$

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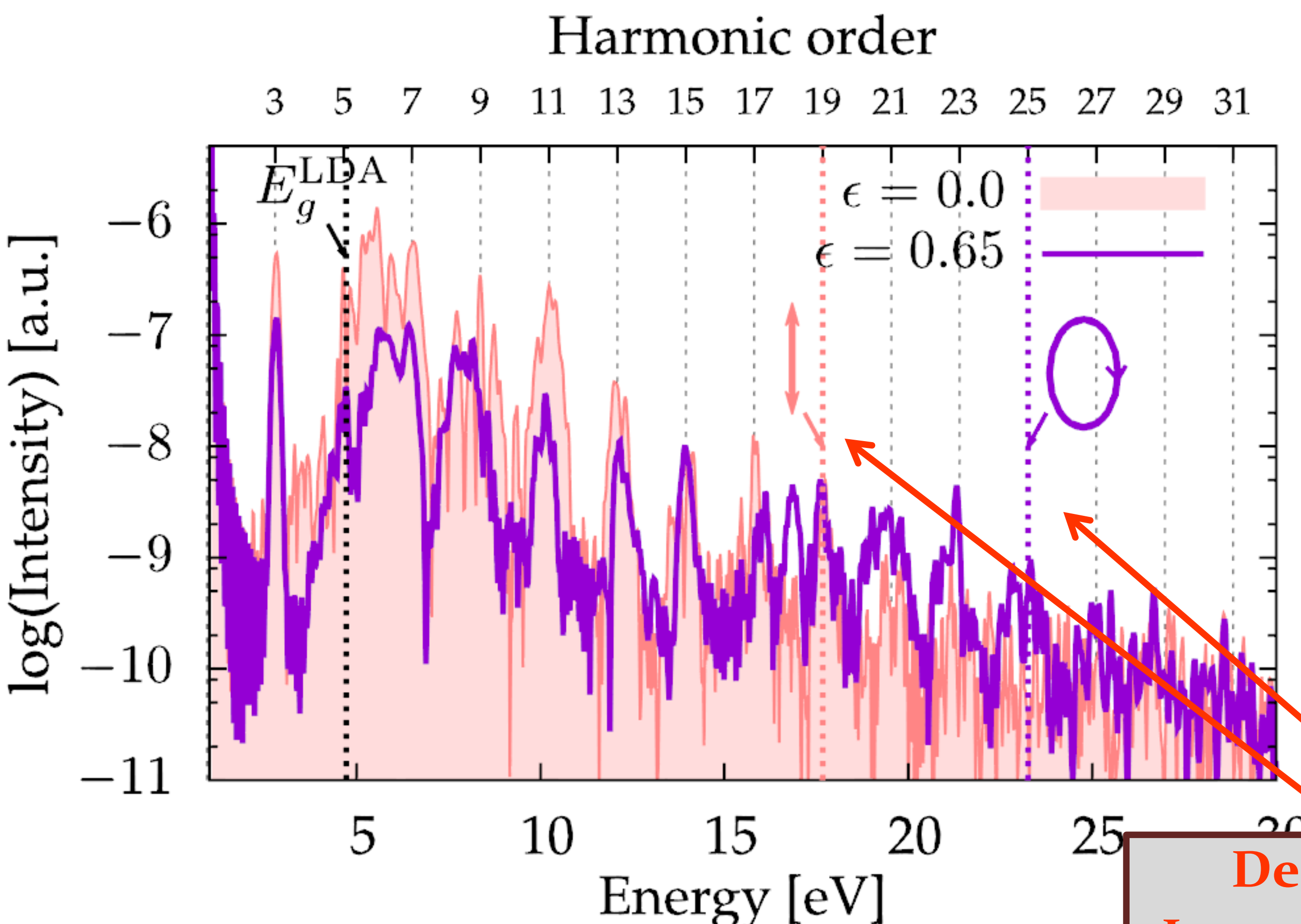
Energy cutoff increase from ellipticity



Bulk MgO
λ=1333nm
I=3x10[12] W/cm²
50fs FWHM

Decrease expected
Instead, increase by 30%

[1] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

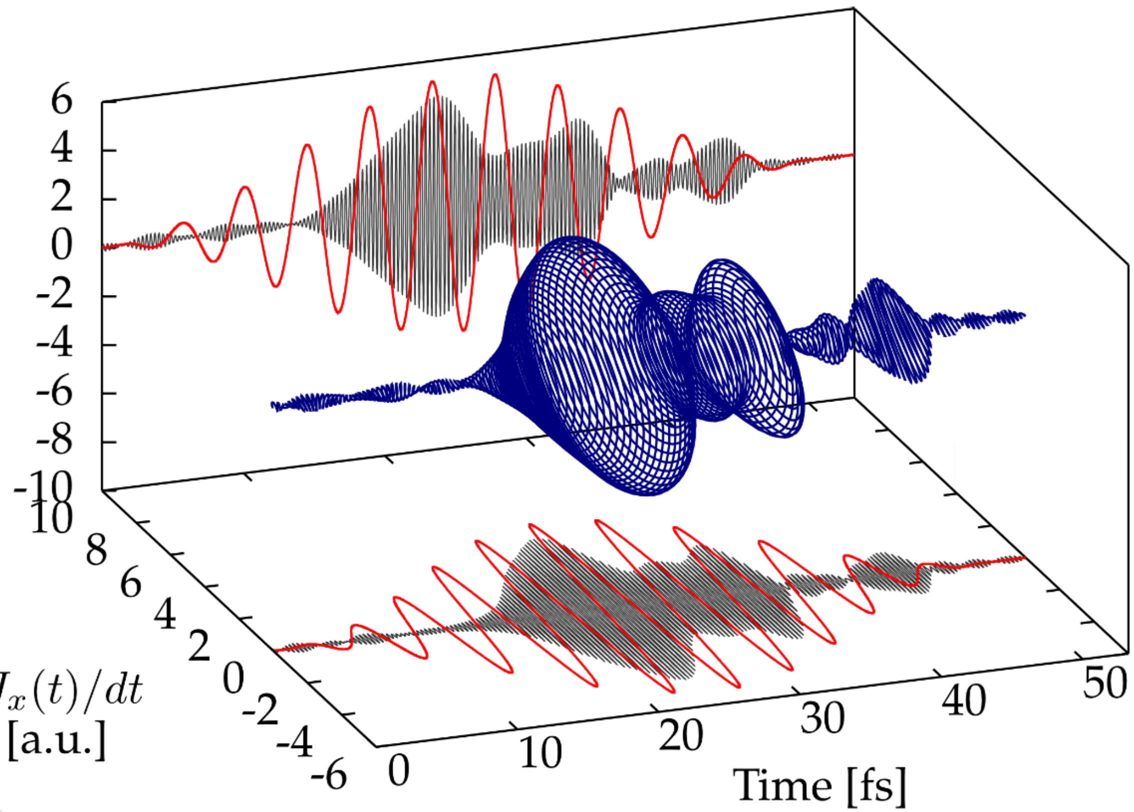
Circularly polarized harmonics from solids

mpsd

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.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

Conclusion

HHG enhanced by inhomogeneity of the electron-nuclei potential

HHG is anisotropic in bulk crystal, even in cubic materials

Possible to suppress interband contribution in favor of HHG yield

Possible to predict the optimal laser polarization, based on the sole knowledge of the crystal's band structure

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

Conclusion

Interband and intraband mechanisms react differently to driver ellipticity

HHG cutoff can be improved by ellipticity

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

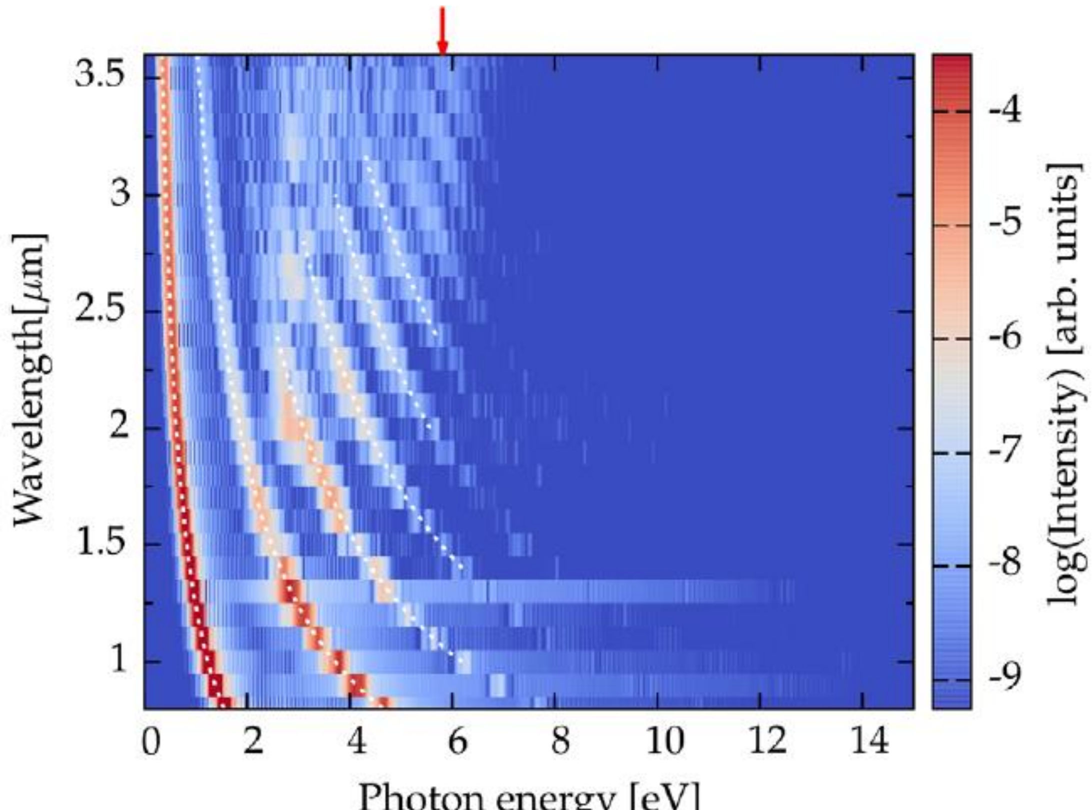
Thank you for your attention

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

[2] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)

Wavelength dependence of the energy cut-off

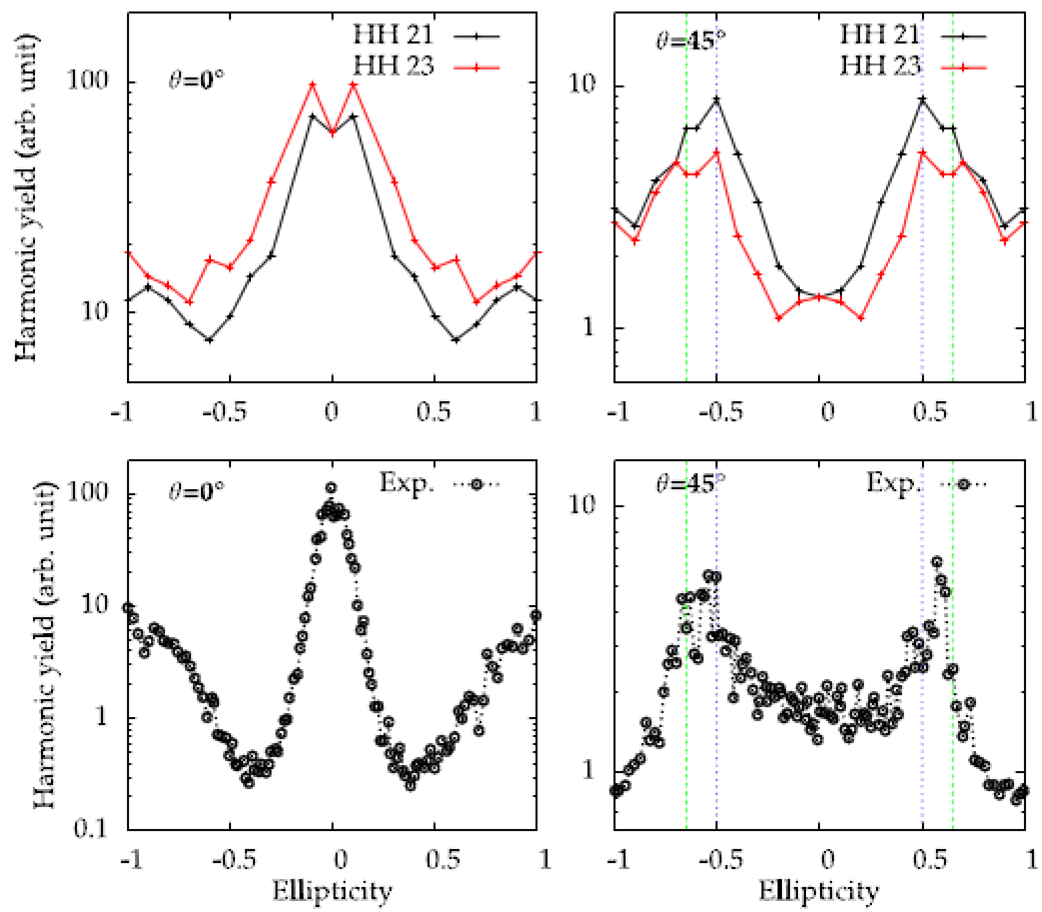
Energy cutoff independent of the wavelength in solids



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

Ellipticity dependence in bulk MgO

Comparison between TDDFT (LDA) and experimental results



[1] N. T.-D. *et al.*, *Ellipticity dependence of high-harmonic generation in solids: unraveling the interplay between intraband and interband dynamics* (submitted)