

Strong-coupling regime in electromagnetic pulses
interacting with carriers in structures and bulk media
AFOSR FA9550-22-1-0182

Strong light-matter (free carrier) interactions in bulk and structured materials.

Miroslav Kolesik

James Wyant College of Optical Sciences
University of Arizona

Quantitative modeling in extreme, solid-state NLO

Tools developed:

SgiSBEs: state of the art SBE solver

MPPE: multi-pulse pulse propagation equations (including “high-contrast” material interfaces)

works using sgiSBEs (each “a first” in modeling of non-resonantly driven materials):

- *Determining the electronic states that contribute most to solid-state high-order harmonic radiation*

M Kolesik, Physical Review A 109 (2), 023503 (2024).

- *Sample-orientation effects in solid-state high-harmonic generation: computational study of GaAs*

M Kolesik, JOSAB, 41, B7-B13, (2024)

- *Numerical discreteness and dephasing in high-harmonic calculations in solids*

M Kolesik, JV Moloney, Physical Review B 108 (11), 115433 (2023).

- *Assessment of tight-binding models for high-harmonic generation in zinc blende materials*

M Kolesik, Optics Letters 48 (12), 3191-3194 (2023).

- *Full-Brillouin-zone calculation of high-order harmonic generation from solid-state media*

J Gu, M Kolesik, Physical Review A 106 (6), 063516 (2022).

works using MPPE

- *Propagation and material-interface effects in HHG from solid-state samples*

M Kolesik, Physical Review A 110, 033512 (2024).

- ***Reflection-geometry propagation effects in the higher-order harmonic radiation from solid-state materials***

M Kolesik, Optics Continuum, to appear.

- ***Full Brillouin zone, multi-band reconstruction of the electronic structure from high-harmonic spectra***

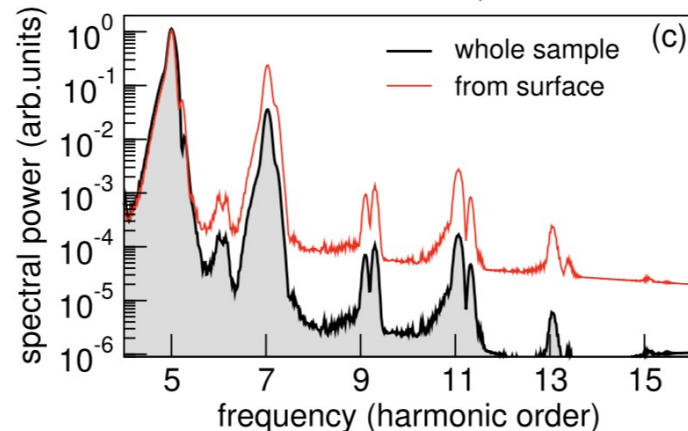
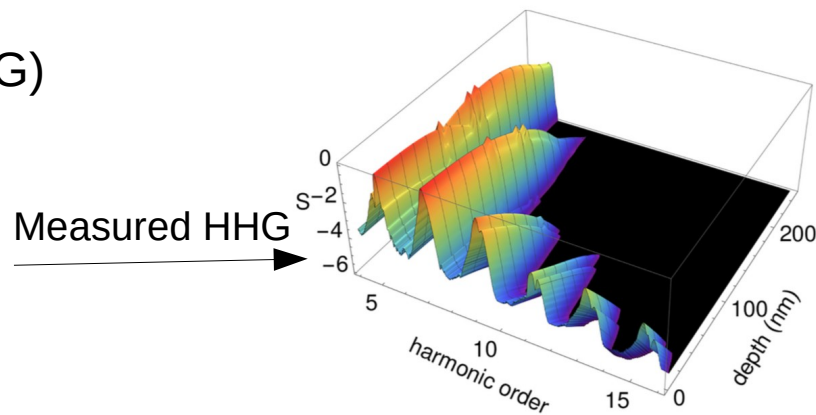
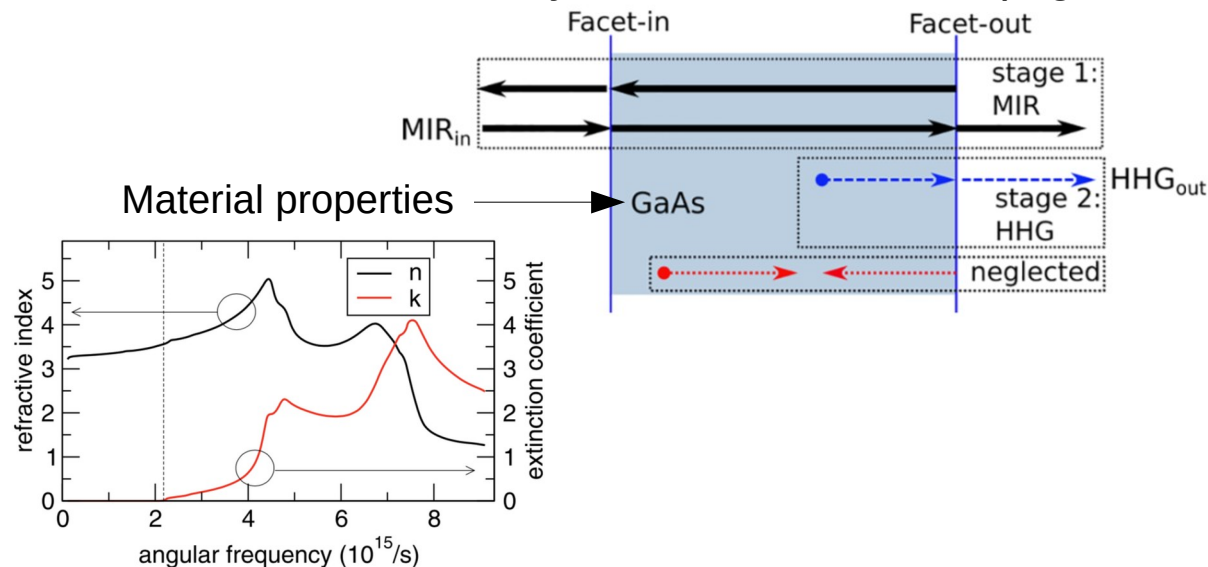
A M Parks, M Kolesik, Optics Express, to appear.

today's focus

Multi-Pulse Propagation Equations: For when uni-directional approximation fails

Sample-thickness effects, free carriers, nonlinearity, interfaces...

Geometry: TRANSMISSION, (e.g. T-HHG)



Take-away:

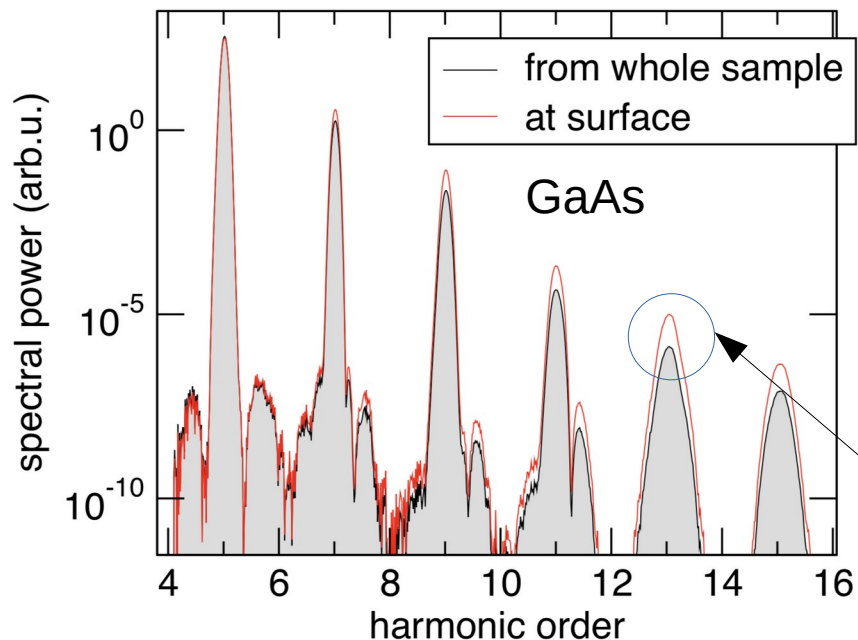
Pronounced propagation effects require large-scale simulations

Reason = bi-directionality + harmonics originate at different depths

Multi-Pulse Propagation Equations: Application to measurements in reflection

Sample-thickness effects due to free carriers, nonlinearity, ...

Experiment geometry: REFLECTION, (R-HHG)



Simulation fully including propagation effects:

- Relevant mechanism = phase mismatch
- Effective sample depth \sim hundreds of nm
- Material-dependent
- Absorption important
- Fresnel reflection less important
- Simple+accurate treatment can be designed

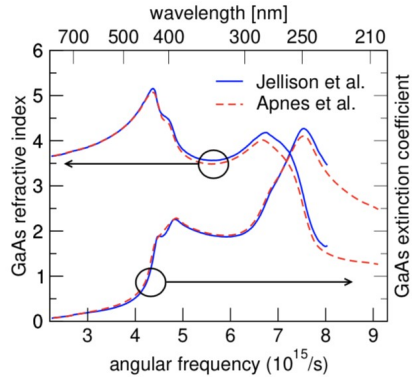
factor ~ 5

“small” difference for practical purposes

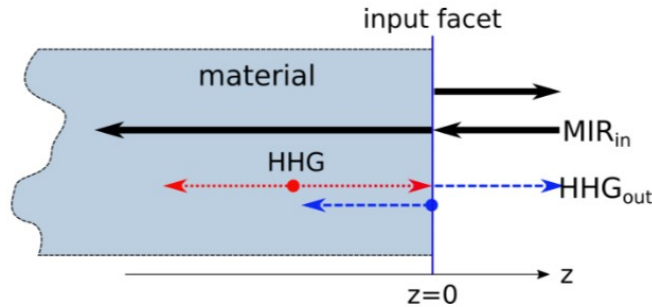
Q: Given the effect is small, is it possible to avoid these very expensive computations (while still capturing all propagation effects)?

Multi-Pulse Propagation Equations: From full simulation to simplified treatment

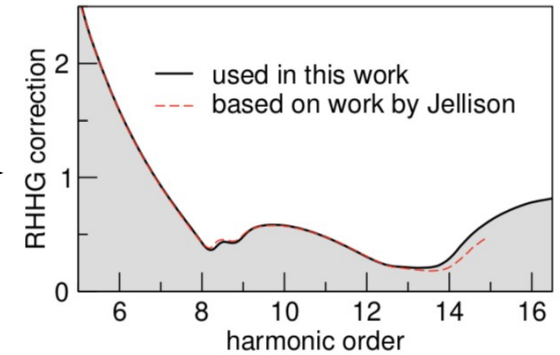
Sample-thickness effects due to free carriers, nonlinearity, ...



Geometry: REFLECTION, (R-HHG)



correction



$$S_{\text{obs}}(\omega) \sim \left| \frac{\omega}{k_z(\omega)} \frac{t(\omega)}{k_z(\omega) + \omega/v_p} \right|^2 S_{\text{surf}}(\omega) \sim \left| \frac{\omega_R [n(\omega_R) + 1] [n(\omega_R) + n(\omega_0)]}{\omega [n(\omega) + 1] [n(\omega) + n(\omega_0)]} \right|^2 S_{\text{surf}}(\omega),$$

Fresnel reflection,
Absorption,
Phase mismatch

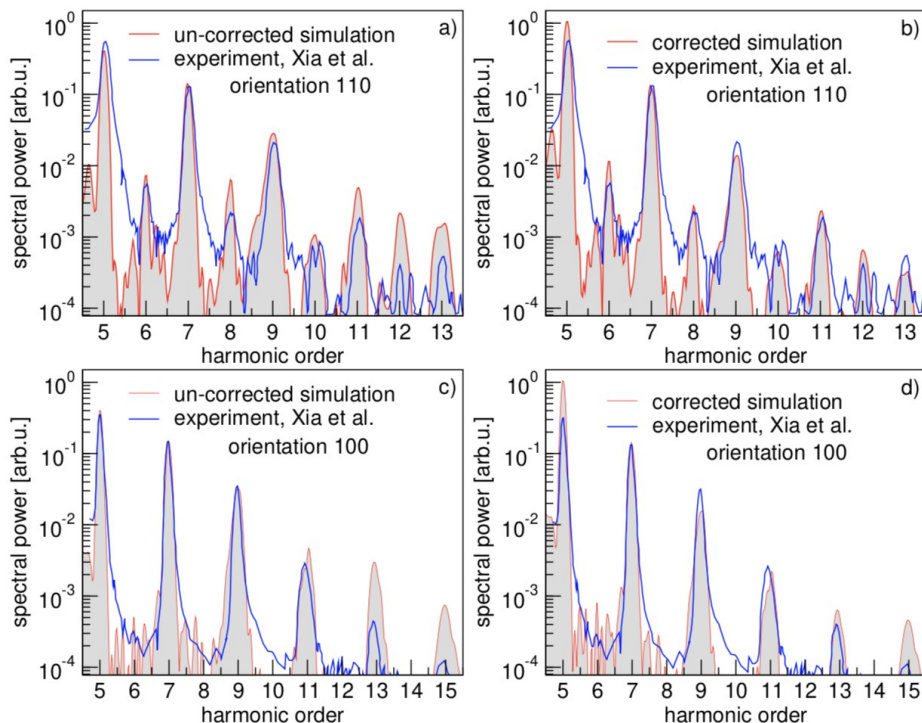
normalized at reference frequency

Inexpensive simulation

Simplified yet accurate treatment of propagation effects in Reflection-HHG

Sample-thickness effects due to free carriers, nonlinearity, ...

Geometry: REFLECTION, (R-HHG)



Simulated-vs-measured HHG:

Modest improvement

after inclusion of propagation effects

$$S_{\text{obs}}(\omega) \sim \left| \frac{\omega}{k_z(\omega)} \frac{t(\omega)}{k_z(\omega) + \omega/v_p} \right|^2 S_{\text{surf}}(\omega) \sim \left| \frac{\omega_R [n(\omega_R) + 1] [n(\omega_R) + n(\omega_0)]}{\omega [n(\omega) + 1] [n(\omega) + n(\omega_0)]} \right|^2 S_{\text{surf}}(\omega),$$

Take-away:

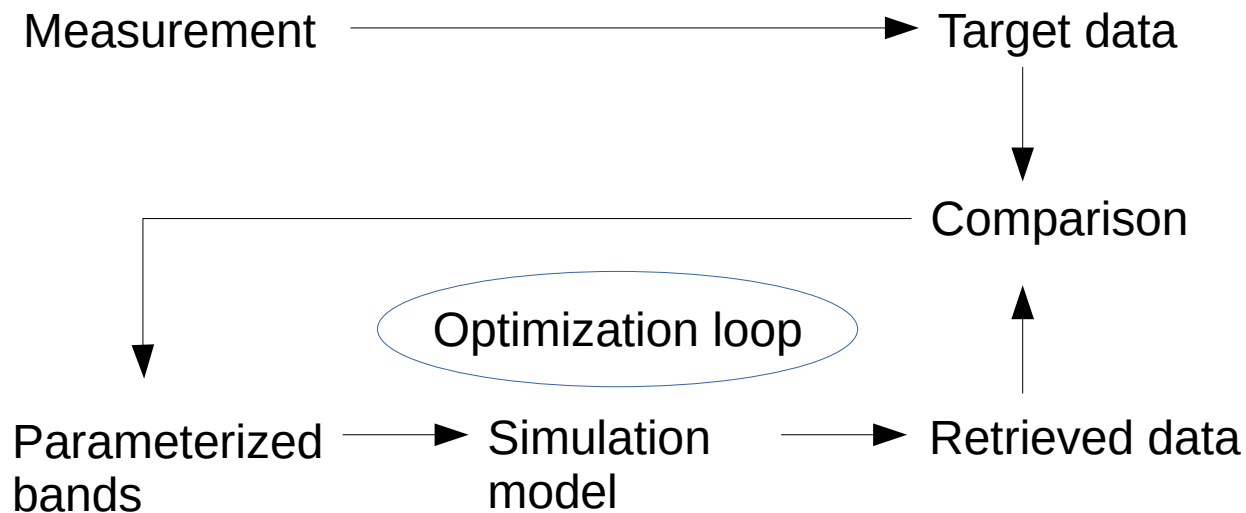
Yes, propagation effects are weak

Yet, important for (future) quantitative comps.

Simple formula + point-simulation

Inexpensive, so no reason not to include

All-optical band-structure reconstruction as an optimization problem



Band-structure reconstruction as an optimization problem

PRL **115**, 193603 (2015)

PHYSICAL REVIEW LETTERS

week ending
6 NOVEMBER 2015



All-Optical Reconstruction of Crystal Band Structure

G. Vampa,^{1,*} T. J. Hammond,¹ N. Thiré,² B. E. Schmidt,² F. Légaré,² C. R. McDonald,¹ T. Brabec,¹
D. D. Klug,³ and P. B. Corkum^{1,3,†}

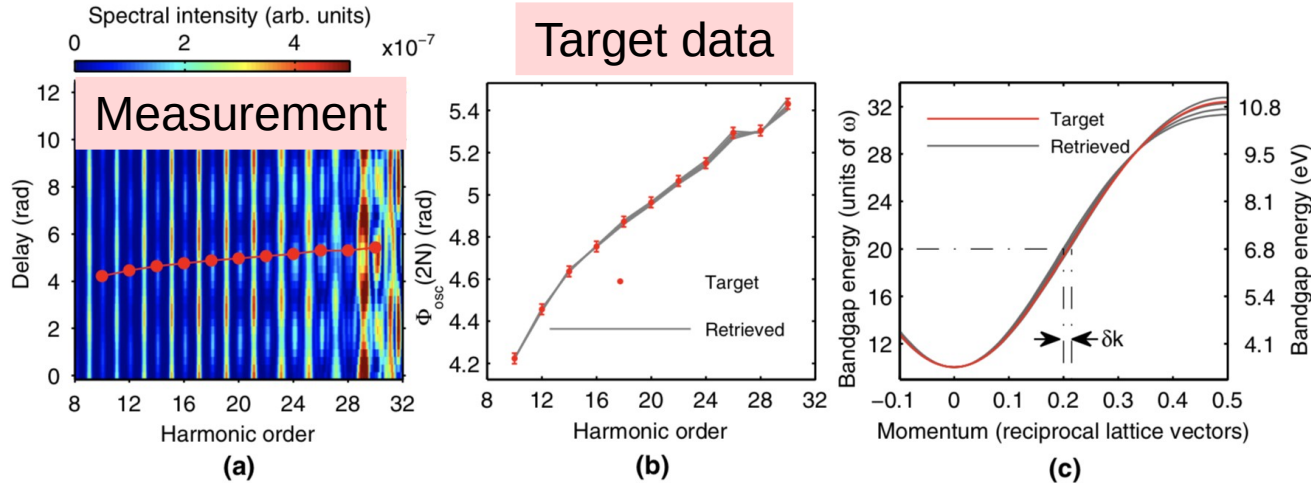


FIG. 2 (color online). Reconstruction of the bands. (a) High harmonic spectrum obtained from the target band structure as a function of delay between fundamental and second harmonic fields. The red line with dots is the optimum phase Φ_{osc} . (b) Target Φ_{osc} (red dots with error bars) compared to those associated with the retrieved band structures (gray lines). (c) Target (red line) and retrieved (gray lines) momentum-dependent band gaps. The Brillouin zone extends up to half of the reciprocal lattice vector. The target band gap is $\varepsilon(k) = \varepsilon_g + 0.139 [1 - \cos(ka)] + 0.011 [1 - \cos(2ka)]$, and approximates that of a ZnO crystal as obtained from *ab initio* calculations. The crystal and laser parameters are defined in Table S2 of the Supplemental Material [19].

Band-structure reconstruction as an optimization problem

PRL **115**, 193603 (2015)

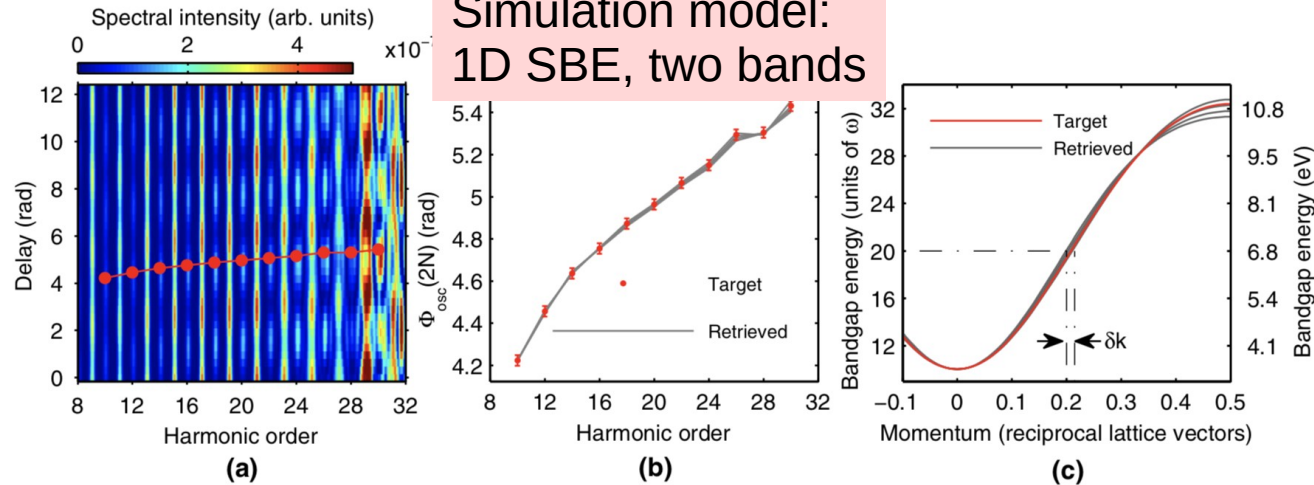
PHYSICAL REVIEW LETTERS

week ending
6 NOVEMBER 2015



All-Optical Reconstruction of Crystal Band Structure

G. Vampa,^{1,*} T. J. Hammond,¹ N. Thiré,² B. E. Schmidt,² F. Légaré,² C. R. McDonald,¹ T. Brabec,¹
D. D. Klug,³ and P. B. Corkum^{1,3,†}

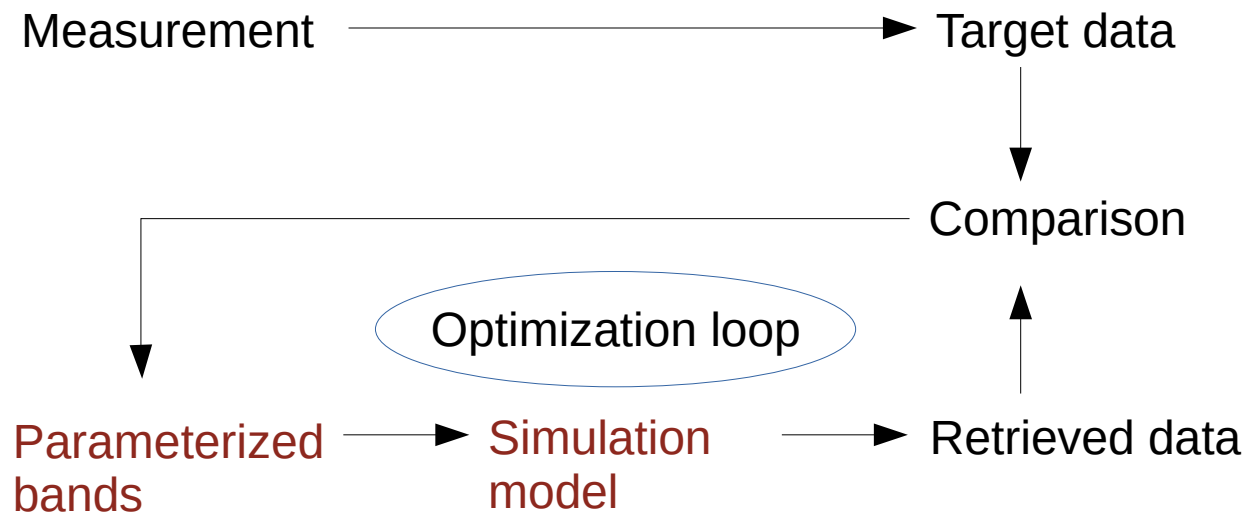


Parameterized
bands

Reconstruction of the bands. (a) High harmonic spectrum obtained from the target band structure as a function of fundamental and second harmonic fields. The red line with dots is the optimum phase Φ_{osc} . (b) Target Φ_{osc} (red dots with error bars) compared to those associated with the retrieved band structures (gray lines). (c) Target (red line) and retrieved (gray lines) bandgap energy vs momentum. The Brillouin zone extends up to half of the reciprocal lattice vector. The target band gap is

$\varepsilon(k) = \varepsilon_g + 0.139 [1 - \cos(ka)] + 0.011 [1 - \cos(2ka)]$, and approximates that of a ZnO crystal as obtained from *ab initio* calculations. The crystal and laser parameters are defined in Table S2 of the Supplemental Material [19].

All-optical band-structure reconstruction as an optimization problem



Most important limitations

Previous approaches, assumptions, approximations, ...

- ▶ Only a subset of processes included (e.g. intra-currents)
- ▶ Only a small set of bands used (mostly two)
- ▶ Only a small “slice” of the Brillouin zone considered (mostly a 1D lineout)
- ▶ Based on semi-classical approximation of HHG (in some form, almost all works)
- ▶ Transition dipole moments assumed to be known and/or simplified (e.g. down to a constant)

Unrealistic: knowledge of dipoles \gg knowledge of energies

Inconsistent: change bands \longrightarrow change dipoles

While avoiding approximation used in previous works, we ask:

- ▶ What kind and how many measurements are needed?
- ▶ How many bands can be reconstructed?
- ▶ Over what part of the Brillouin zone?

Our approach: Surrogate-model for band-structure reconstruction

Rather than the band-energies, parameterize a surrogate Hamiltonian:

$$H(\mathbf{k}, \{p_i\})$$

- ▶ Strictly enforce the material symmetry
- ▶ Multi-band
- ▶ Full Brillouin zone
- ▶ Transition-dipole moments + Berry connections (if desired) **obtained as by-products**

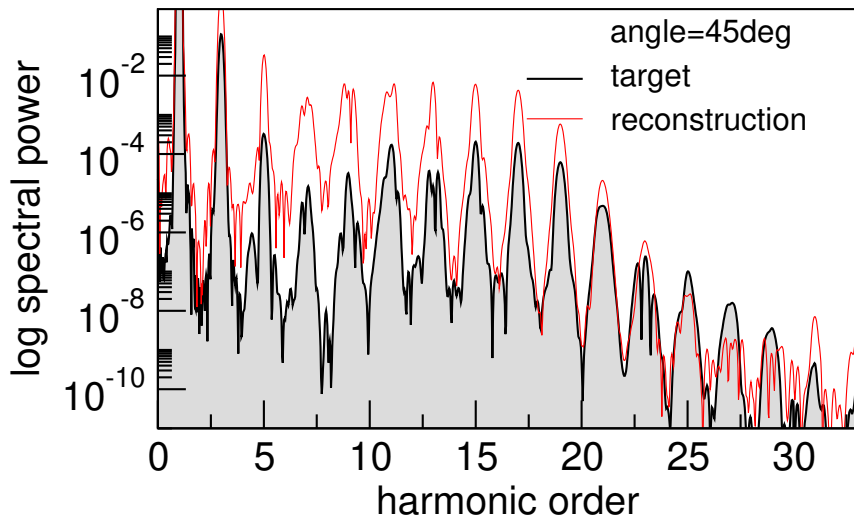
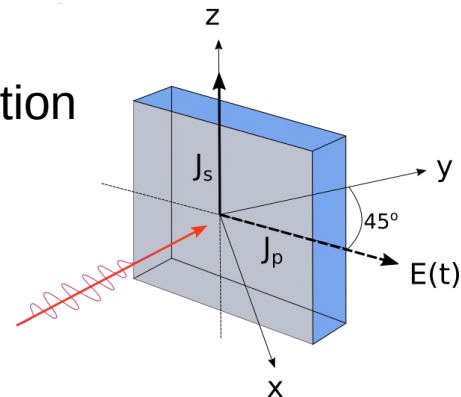
This work:
non-resonantly excited
GaAs, ZnSe, Si, ...
TB-models with 10, 20 bands
7 – 13 parameters p_i

Requires a large-scale simulation to optimize $\{p_i\}$

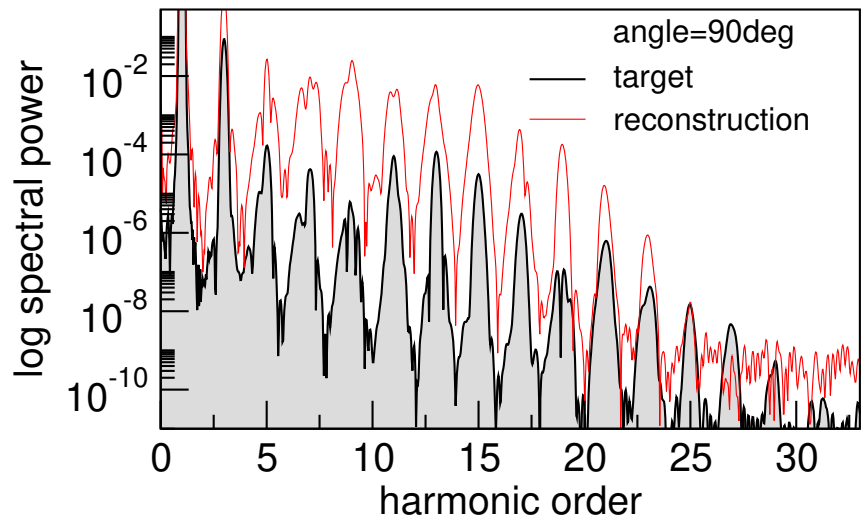
Surrogate Hamiltonian for band-structure reconstruction: Measurement inputs

Input data (= target of the optimization)

A **single pair of HHG spectra**, rotated (100) sample, in reflection
Silicon example:



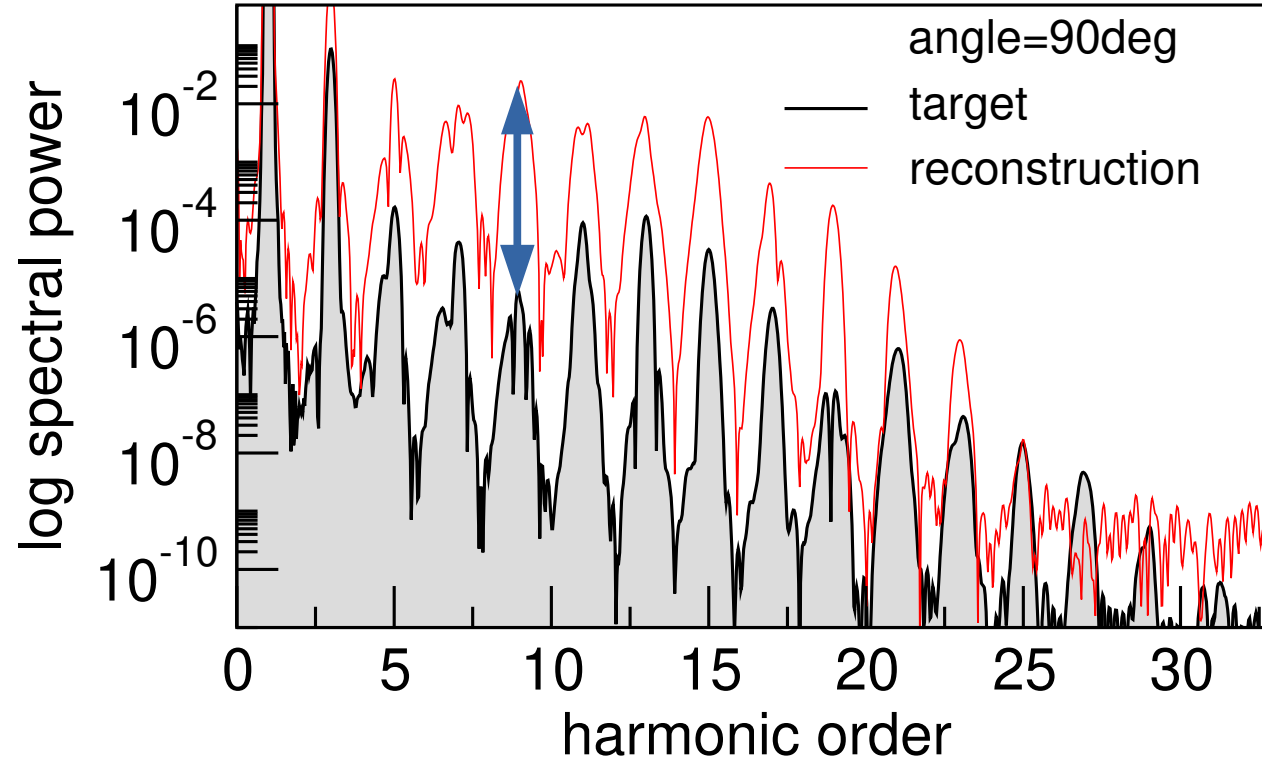
Here, with no optimization,
Reconstruction = first guess



Surrogate Hamiltonian for band-structure reconstruction: Fitness function

Choice of the fitness function:

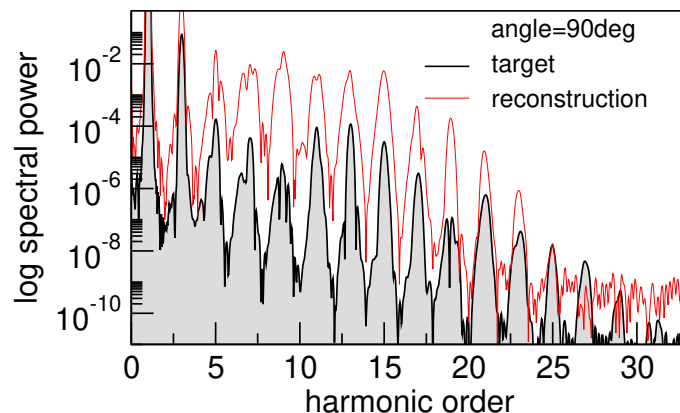
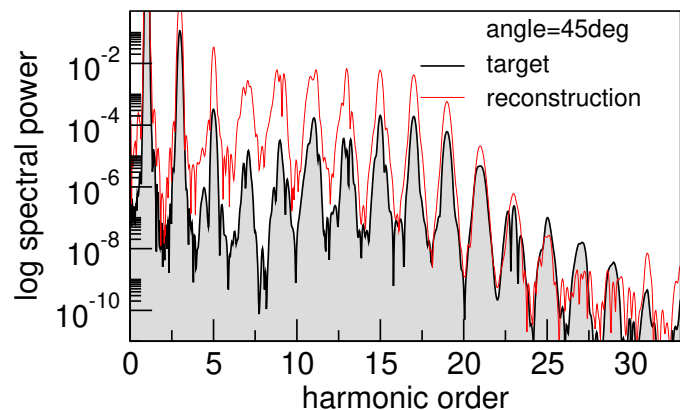
Average absolute difference between the target and the reconstruction spectra in log scale



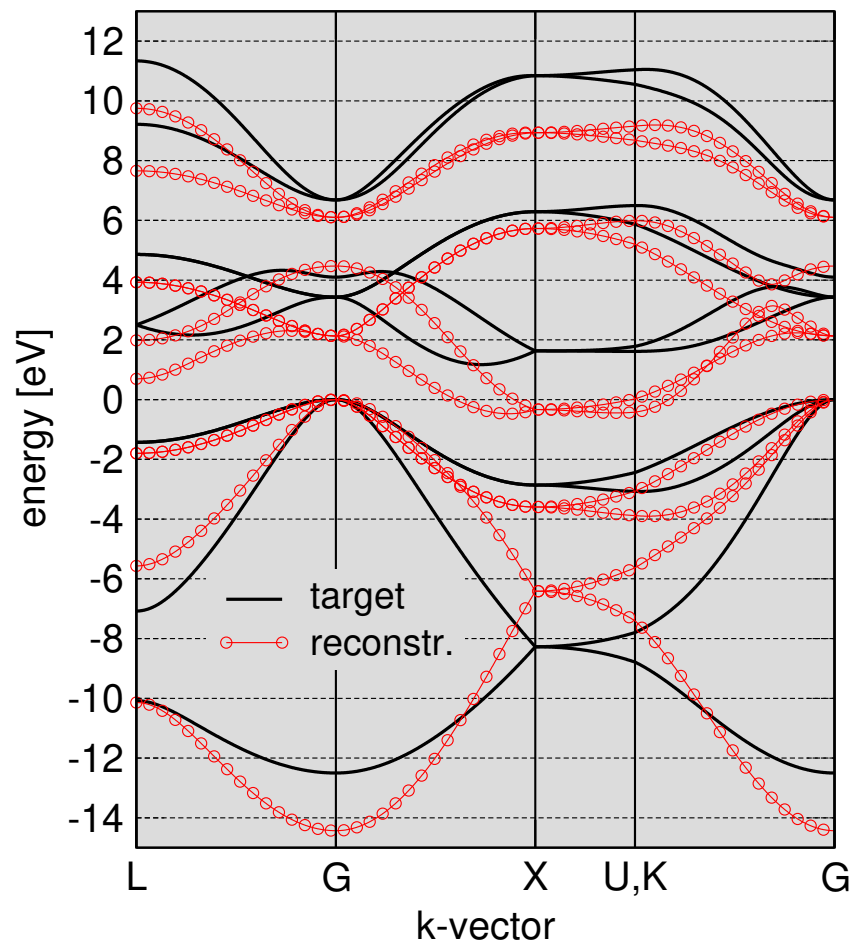
Surrogate Hamiltonian for band-structure reconstruction: Initial guess

Initial surrogate model:

Here: Randomized target parameters



Initial guess (symbols) vs target (line)



Surrogate Hamiltonian for band-structure reconstruction: Fitness landscape

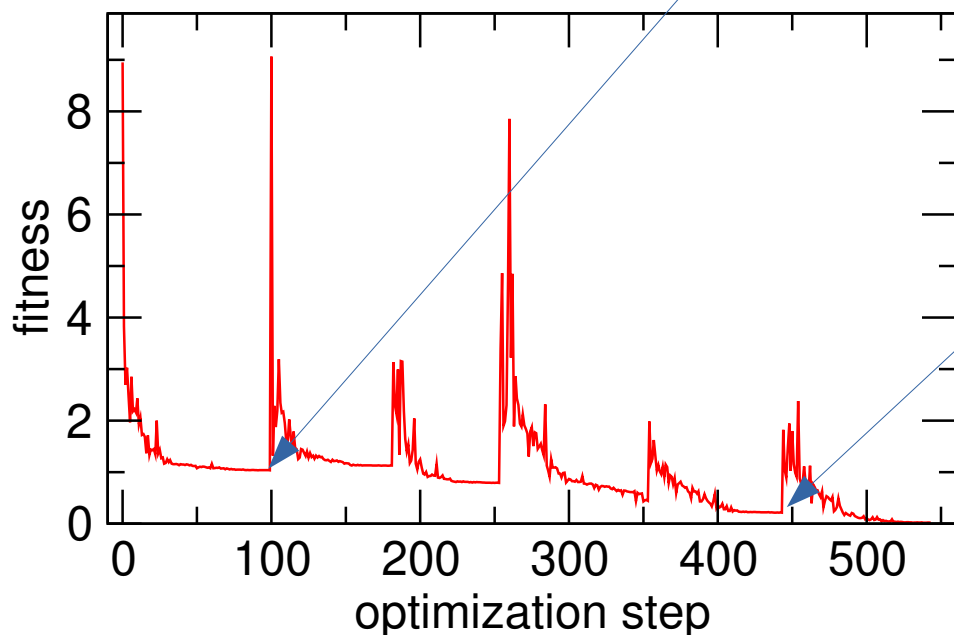
Fitness landscape is complex:

Not surprising: “every” reasonable surrogate mode produces “good-looking HHG”

Challenging: Large number of “false” solutions

Optimization algorithm:

1. Simplex
2. Stochastic escapes from stalled solutions



Silicon example

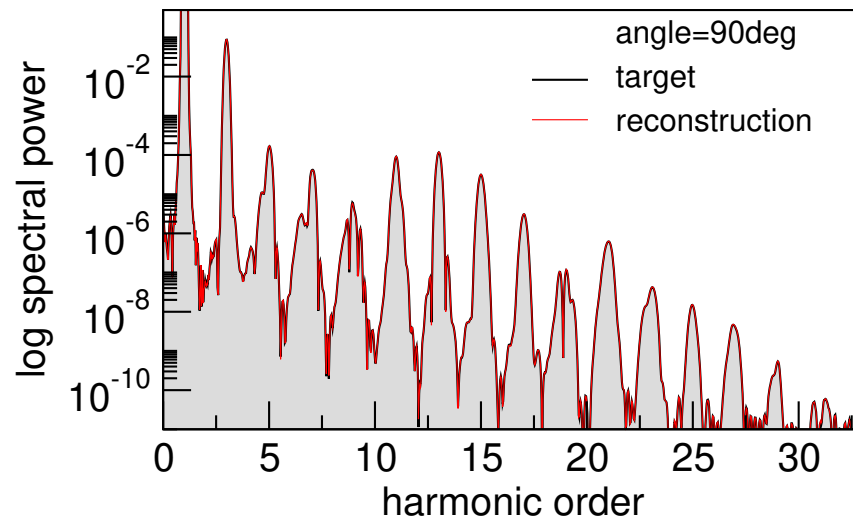
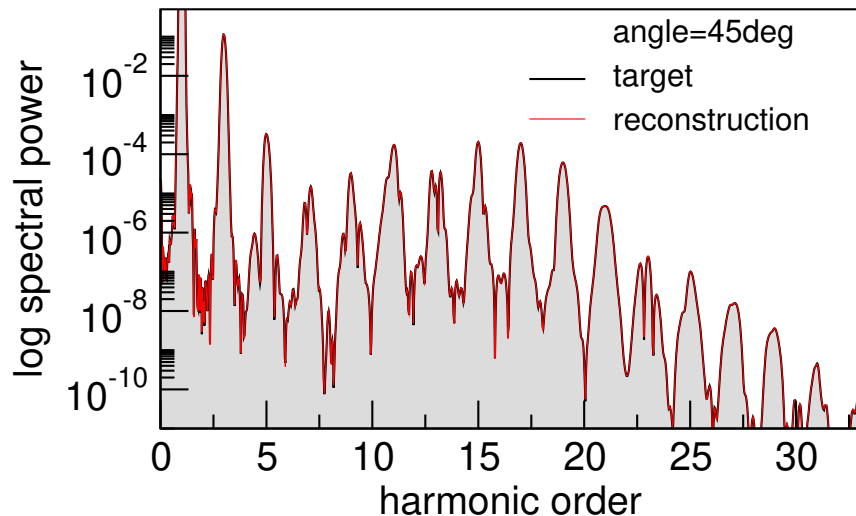
n	f	p_1	p_2	p_3	p_4	p_5	p_6	p_7	p_7
t	0.0	-4.200	1.715	6.685	-8.300	1.715	4.575	5.729	5.375
*	8.948	-4.979	1.065	6.102	-9.451	1.065	4.661	2.892	4.538
0	1.036	-3.782	1.853	4.747	-9.003	1.853	5.373	5.000	4.787
1	1.128	-3.080	1.843	5.039	-8.088	1.844	5.395	4.891	5.105
2	0.793	-2.887	1.742	5.411	-8.024	1.742	5.060	5.560	5.008
3	0.445	-2.755	1.718	6.418	-6.843	1.718	4.649	5.838	5.270
4	0.214	-3.022	1.715	5.921	-7.111	1.716	4.594	5.845	5.139
5	0.021	-3.817	1.714	6.717	-7.909	1.714	4.575	5.708	5.464
t	0.0	-4.200	1.715	6.685	-8.300	1.715	4.575	5.729	5.375

t target
* initial guess
0..5 stalled solutions

Surrogate Hamiltonian for band-structure reconstruction: Convergence

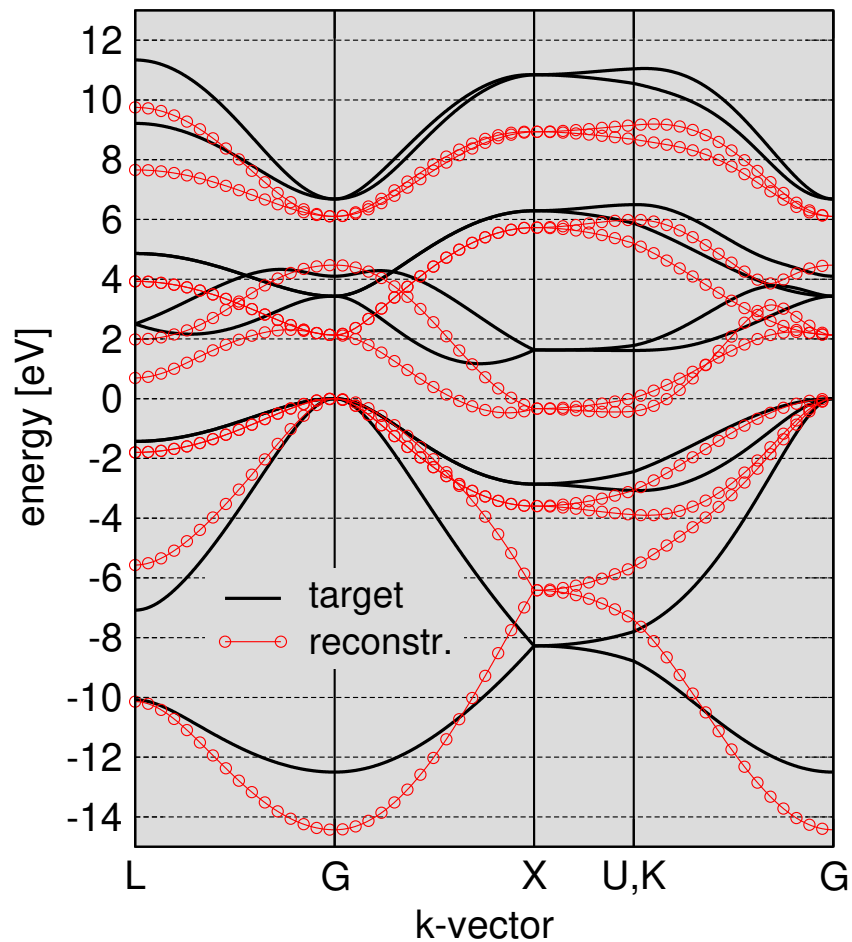
Convergence to the target data:

Nearly perfect (perfect possible when the ideal solution does exist)

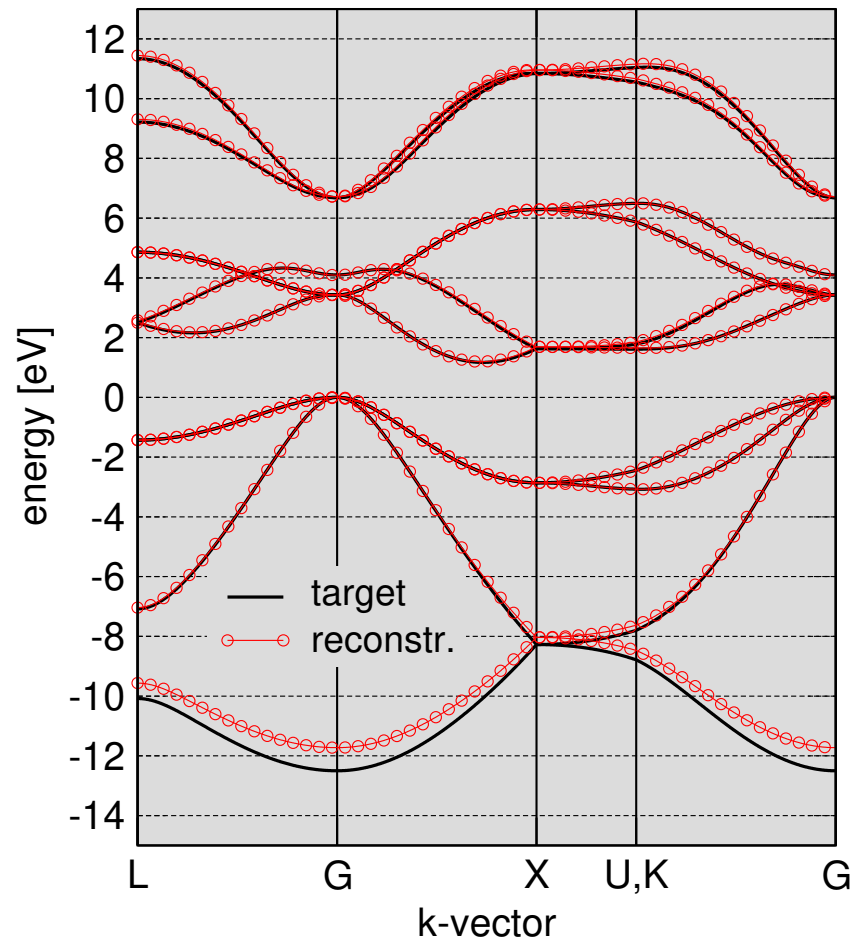


Band-structure reconstruction as an optimization problem: Convergence

Initial (Silicon example)

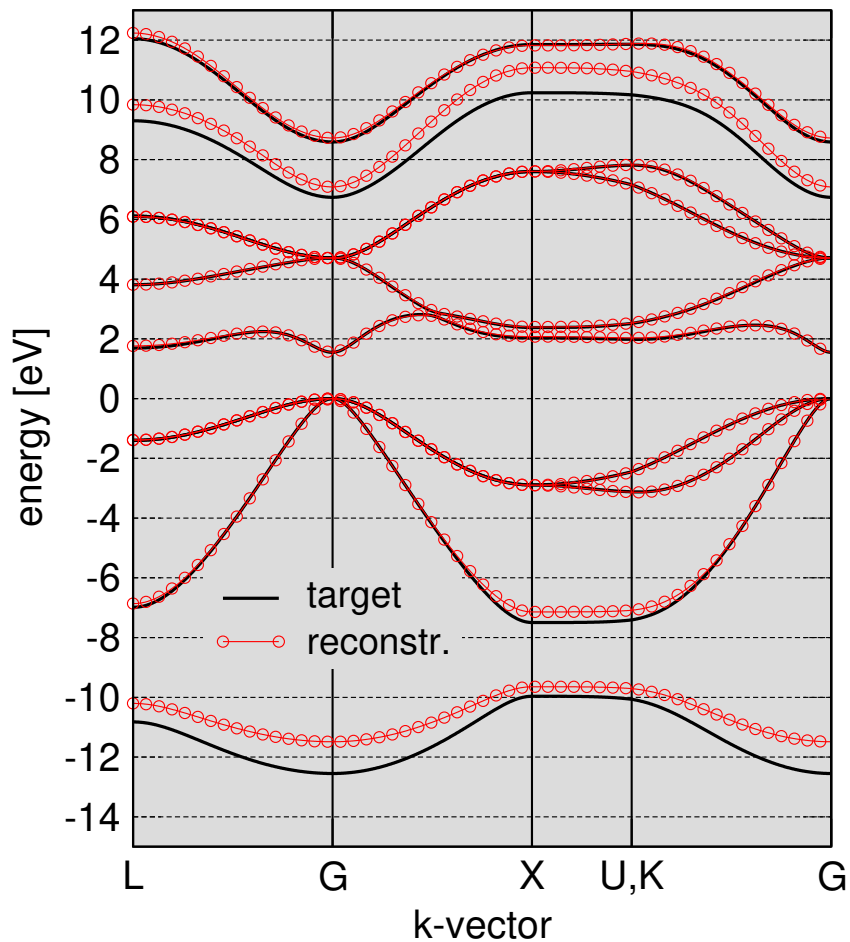


Reconstructed

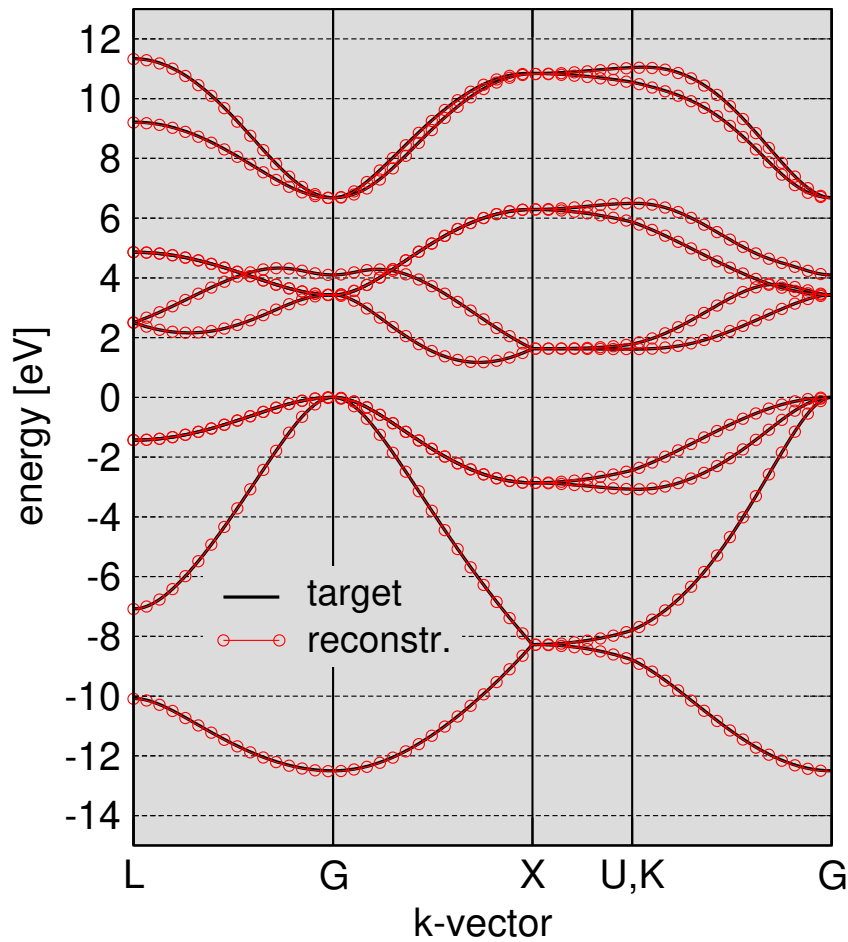


Band-structure reconstruction as an optimization problem: Convergence

Accepted solution (GaAs example)



Can reach the true solution
(further optimization, Silicon)

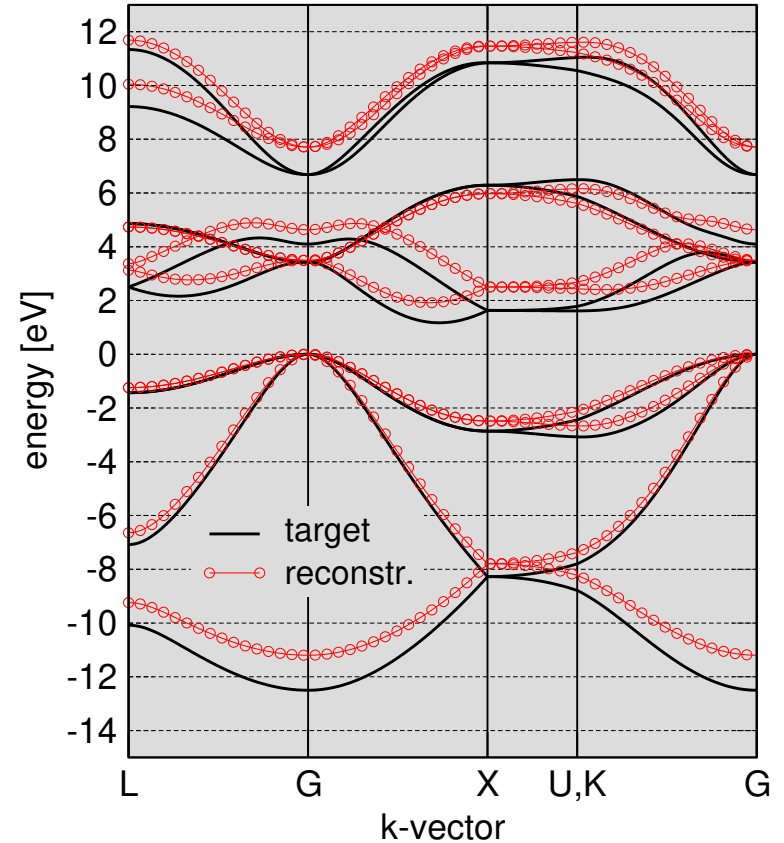
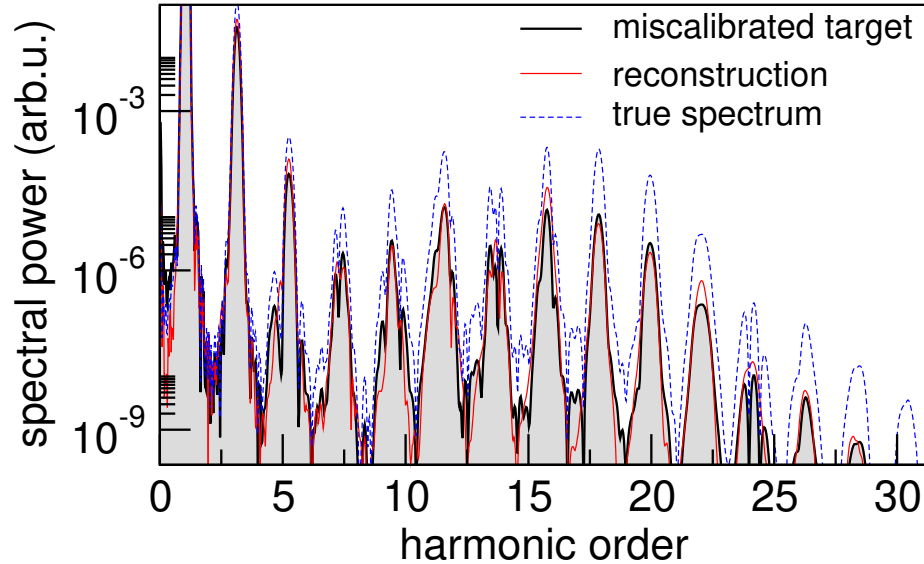


It works, but is this (first test of the method) realistic?

Impacts of target-data quality

Testing robustness:

Assume HHG spectrum is mis-calibrated

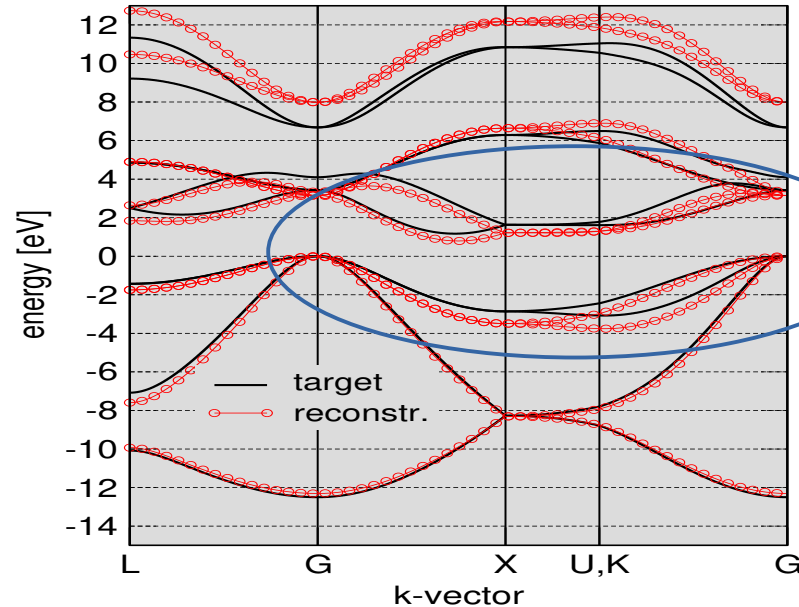


Take away: accurate spectral calibration necessary

Impacts of target-data quality

Testing robustness:

Assume error in (excitation) peak amplitude (Si example)



Peak amplitude 30% off

Take away:

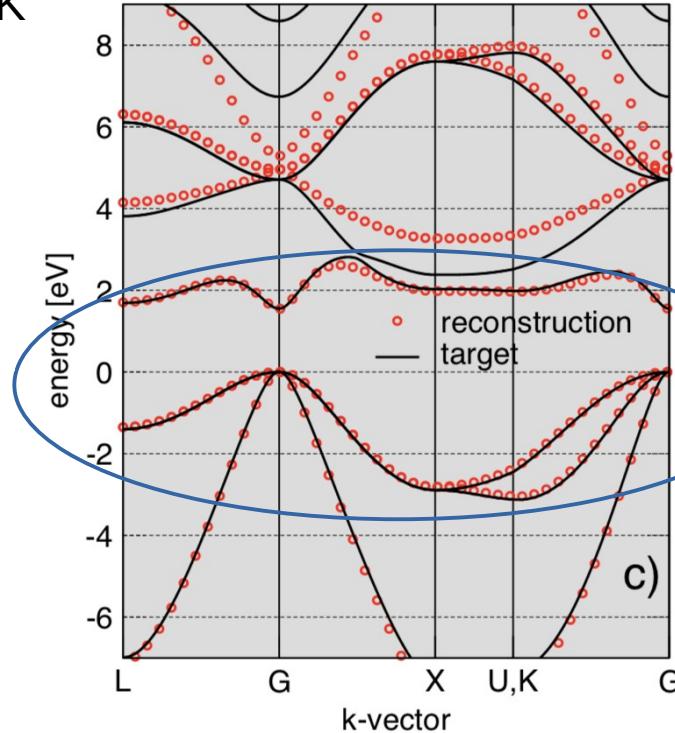
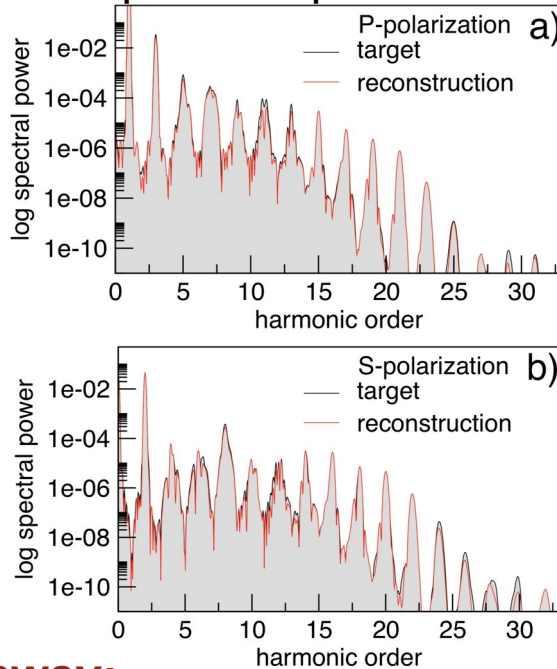
Surrogate-model approach is surprisingly (?) robust w.r.t. characterization of the excitation pulse

Impacts of target-data quality

Testing robustness:

Assume error in de-phasing rate: Target $T_2 = 5\text{fs}$ vs Surrogate model $T_2 = 8\text{fs}$

Spectra reproduced OK



Important bands OK

Take away:

De-phasing rate can be treated as yet another parameter to optimize
Reasonable first guess needed --- Obtain HHG-governing bands right

Conclusions

Band-structure reconstruction from a single-pulse, single-color excited HHG:

- ▶ What kind and how many measurements are needed?

Reflection-HHG, few well-chosen spectra are IN PRINCIPLE sufficient

- ▶ How many bands can be reconstructed?

Minimum: Multiple energetically “connected” bands surrounding the gap

- ▶ Over what part of the Brillouin zone?

Entire Brillouin zone, with symmetry properties enforced by surrogate Hamiltonian

Ours is still a proof-of-a-concept study, but results are extremely encouraging

Hoping to stimulate renewed interest in all-optical reconstruction of band-structure

Conclusions

Band-structure reconstruction from a single-pulse, single-color excited HHG:

- ▶ What kind and how many measurements are needed?

Reflection-HHG, few well-chosen spectra are IN PRINCIPLE sufficient

- ▶ How many bands can be reconstructed?

Minimum: Multiple energetically “connected” bands surrounding the gap

- ▶ Over what part of the Brillouin zone?

Entire Brillouin zone, with symmetry properties enforced by surrogate Hamiltonian

Going forward...

- ▶ Working with real experimental data:

Experimental accuracy + spectral calibration = important issues

Calibration-immune schemes

- ▶ Richer material models (SOC, many-body, band-renormalization, two-D systems,...)

