High-Order Harmonic Generation in Solids

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Day 1

9:30-10:10 *Probing and generating high harmonics with electron beams.*
Javier García de Abajo (invited).

10:10-10:50 *High harmonic spectroscopy of strongly correlated and topological materials.*
Rui Emanuel Ferreira da Silva (invited).

10:50-11:10 *Macroscopic high harmonic generation in single-layer graphene dominated by a phase-matched ring.* **Roberto Boyero-García,** Óscar Zurrón-Cifuentes, Luis Plaja, Carlos Hernández-García.

11:10-11:30 *Strong-field-driven dynamics and high-harmonic generation in interacting one dimensional systems.* **Joel D. Cox**, Sandra de Vega, Fernando Sols, F. Javier García de Abajo.

11:30-11:50 *Lightwave topology for strong-field valleytronics: inducing, controlling and reading the valley pseudospin on a sub-laser-cycle timescale.* **Á. Jiménez-Galán**, Rui E.F. Silva, O. Smirnova, M. Ivanov.

11:50-12:10 *Helicity flip of harmonics from finite and infinite topological nanoribbons.*
Christoph Jürß, Daniel Moos, Dieter Bauer.

12:10-12:30 *Cluster approach to attosecond photoelectron emission from dichalcogenides.*
Marcelo J. Ambrosio, Etienne Plesiat, Piero Decleva, Pedro M. Echenique, Ricardo Díez Muiño, Fernando Martín.

Day 2

9:30-10:10 *Attosecond metrology of phase-coherent multi-PHz currents in bulk solid.*
Manish Garg (invited), M. Zhan, H.Y. Kim, H. Lakhotia, E. Goulielmakis.

10:10-10:30 *Origin of strong-field-induced low-order harmonic generation in amorphous quartz.*
P. Jürgens, B. Liewehr, B. Kruse, C. Peltz, D. Engel, A. Husakou, T. Witting, M. Ivanov, M. J. J. Vrakking, **T. Fennel**, A. Mermillod-Blondin.

10:30-10:50 *Attosecond transient absorption spectroscopy in condensed matter systems.*
G. Cistaro, L. Plaja, F. Martín, A. Picón.

10:50-11:10 *High order harmonic generation spectroscopy of ZnTe.*
Davide Faccialà, Aditya Pusala, Prabhash Prasannan Geetha, Mikayel Musheghyan, Gabriele Crippa, Anna G. Ciriolo, Michele Devetta, Andreas Assion, Eugenio Cinquanta, Caterina Vozzi, Salvatore Stagira.

11:10-11:30 *Signatures of electronic Talbot revivals in HHG from a one-dimensional crystal.* **Ana García-Cabrera**, Carlos Hernández-García, Luis Plaja.

11:30-11:50 *Following Phonon Dynamics in Solids Using High-Harmonic Spectroscopy.* **T. T. Luu**, F. Lengers, M. Molitor, D. Wigger, D. E. Reiter, T. Kuhn, H. J. Wörner.

11:50-12:10 *Ab initio study of high-order harmonics and isolated attosecond pulses from a solid target.* **Zahra Nourbakhsh**, Nicolas Tancogne-Dejean, Oliver D. Mücke, Hamed Merdji, Angel Rubio.

12:10-12:30 *Mechanism for high-order harmonic generation in graphene and carbon nanotubes.* **Óscar Zurrón-Cifuentes**, Roberto Boyero-García, Carlos Hernández-García, Antonio Picón, Luis Plaja.

Posters

1. *A comprehensive study of momentum matrix elements for Bloch states using a local orbital basis.* **J. J. Esteve-Paredes**, J. J. Palacios.
2. *Low order harmonics of an organic polymer.* **Franziska Fennel,** Stefan Lochbrunner.
3. *Beyond-Fröhlich theory of large polarons in anharmonic solids*. **Matthew Houtput**, Jacques Tempere.
4. *Effects of tunnel ionization on non-linear response of solids*. **Benjamin Liewehr**, Steven Lormuß, Thomas Fennel.
5. *Theory of high-harmonic generation in topological insulators*. **Daniel Moos**, Christoph Jürß, Dieter Bauer.
6. *High-energy mid-IR strong-field source for high-order harmonic generation in solids*. **Aditya Pusala**, Prabhash Prasannan Geetha, Davide Faccialà, Mikayel Musheghyan, Gabriele Crippa, Anna G. Ciriolo, Michele Devetta, Andreas Assion, Eugenio Cinquanta, Salvatore Stagira, Caterina Vozzi.
7. *Frequency Upconversion of Quadruple-Gaussian Laser Beams by Excitation of Overtones in InSb with Linearly Increasing Doping Profile.* **Naveen Gupta**.

Probing and generating high harmonics with electron beams

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We will start by reviewing recent advances in high-harmonic generation (HHG) in solids and discuss graphene as a material that is particularly suited to produce strong nonlinear response at mid-infrared fundamental frequencies. Experiments [1] and theoretical analyses [2] will be discussed in a tutorial fashion, as well as some physical limits in the achievable nonlinear energy conversion. In general, materials with highly non-parabolic electron dispersion offer great potential for HHG at those excitation energies, which we will illustrate with a simple order-of-magnitude estimate applicable to a vast range of materials. We will also discuss the ability of electron beams to interact with nonlinear fields and probe them with high efficiency to obtain insight into the nonlinear optical response at the nanometer scale in combination with ultrafast optical illumination [3]. We will conclude by examining the prospects for single [4] and multiple low-energy free electrons to trigger high-harmonic response in nanomaterials.

References

[1] M. Baudisch, A. Marini, J. D. Cox, T. Zhu, F. Silva, S. Teichmann, M. Massicotte, F. H. L. Koppens, L. S. Levitov, F. J. García de Abajo, and J. Biegert, Nature Communications **9**, 1018 (2018).
[2] J. D. Cox and F. J. García de Abajo, Accounts of Chemical Research **52**, 2536-2547 (2019).
[3] A. Konečná, V. Di Giulio, V. Mkhitaryan, C. Ropers, and F. J. García de Abajo, ACS Photonics **7**, 1290-1296 (2020).
[4] J. D. Cox and F. J. García de Abajo, Nano Lett., DOI: 10.1021/acs.nanolett.0c00538.

High harmonic spectroscopy of strongly correlated and topological materials

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The recent discovery of high harmonic generation in solids [1], merging the fields of strong field and condensed matter physics, opened the door for the direct observation of Bloch oscillations [1], all-optical reconstruction of the band structure [2] and direct observation of the influence of the Berry curvature in the optical response [3]. In this work, we will focus on high harmonic generation in strongly correlated and topological materials. First, I will show how high harmonic spectroscopy can be used to induce and time resolve insulator-to-metal transitions in strongly correlated materials, using the Hubbard model [4]. I will further demonstrate how high harmonic spectroscopy can be used to identify topological phases of matter and how the Berry curvature leaves its fingerprint in the nonlinear optical response of the material [5]. At last, I will show how the use of Wannier orbitals can be useful in the calculation of the nonlinear optical response of solids [6].

References

[1] S. Ghimire et al., Nature Physics, **7** 138-141 (2011)
[2] G. Vampa et al, Physical Review Letters, **115** 193603 (2015)
[3] H. Liu et al, Nature Physics, **13** 262-265 (2017)
[4] R. E. F. Silva, Nature Photonics, **12** 266-270 (2018)
[5] R. E. F. Silva, Nature Photonics, **13** 849-854 (2019)
[6] R. E. F. Silva, Physical Review B, **100** 195201 (2019)

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Strong-field-driven dynamics and high-harmonic generation in interacting one dimensional systems

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High-harmonic generation (HHG) is a striking example of a nonlinear optical process and its ability to spectrally and temporally disperse intense laser light. While solid-state HHG is anticipated to pave way for compact ultraviolet and attosecond light sources, it relies on enormous light-matter interaction strengths that are difficult to attain in nano-optical systems with inherently small volumes. Here we argue that the incident light intensity required to trigger HHG can be dramatically reduced by engineering synergies between electronic band structure and collective optical resonances. We demonstrate this principle in one-dimensional (1D) solid-state systems, starting from an idealized model of electrons in finite linear atomic chains governed by the Su-Schrieffer-Heeger (SSH) tight-binding Hamiltonian—driven by an intense ultrashort pulse, we simulate electron dynamics by solving the single-particle density matrix self-consistently, incorporating electron-electron interactions that give rise to collective optical resonances (i.e., plasmons) [1]. The SSH chain is readily tuned between metallic, insulating, and topologically insulating phases, thus constituting a simple but powerful tool to probe the effect of band structure on HHG yield in combination with light-concentrating plasmonic modes introduced through charge carrier doping. The conclusions drawn from the idealized model are demonstrated to hold for realistic carbon-based structures such as graphene nanoribbons and carbon nanotubes [1,2], which offer an intrinsically anharmonic response to light while supporting long-lived and electrically tunable plasmon resonances. The present findings can be straightforwardly extended to optimize existing platforms for HHG or to identify new solid-state alternatives in the context of nonlinear plasmonics.

References

[1] S. de Vega, J. D. Cox, F. Sols, and F. J. García de Abajo, Phys. Rev. Res. **2**, 013313 (2020).
[2] J. D. Cox, A. Marini, and F. J. García de Abajo, Nat. Commun. **8**, 14380 (2017).

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Lightwave topology for strong-field valleytronics: inducing, controlling and reading the valley pseudospin on a sub-laser-cycle timescale

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Modern light generation technology offers extraordinary capabilities for sculpting light pulses, with full control over individual electric field oscillations within each laser cycle [1]. These capabilities are at the core of lightwave electronics -- the dream of ultrafast lightwave control over electron dynamics in solids, on a few-cycle to sub-cycle timescale, aiming at information processing at tera-Hertz to peta-Hertz rates. At the same time, quantum materials [2] encompass fascinating properties such as the possibility to harness extra electronic degrees of freedom, e.g., the valley pseudospin [3]. Previous works have established optical initialization of the valley pseudospin via resonant circularly-polarized pulses [4], and more recently, via few-cycle non-resonant pulses [5]. Still, manipulation and reading of the valley degree of freedom on timescales shorter than valley depolarization remains a crucial challenge [4].

Bringing the frequency-domain concept of topological Floquet systems to the few-femtosecond time domain, I will present an all-optical, non-resonant approach to coherently induce, manipulate and read the valley pseudospin in graphene-like monolayers by controlling the sub-cycle structure of non-resonant driving fields and using the imprint of the Berry curvature on the high harmonic generation spectrum [6,7]. Such tailored fields modify the laser-driven band structure on a sub-laser cycle timescale, opening the way to all-optical ultrafast control of topological properties in quantum materials [8].

References

[1] F. Krausz et al., Rev. Mod. Phys., **81** 163 (2009).
[2] A.H. Castro Neto et al., Rev. Mod. Phys., **81** 109 (2009).
[3] S.A. Vitale et al., Small, **14** 1801483 (2018).
[4] D. Xiao et al., Phys. Rev. Lett., **99** 236809 (2007).
[5] S.A. Oliaei Motlagh et al., Phys. Rev. B **100** 115431 (2019).
[6] Á. Jiménez-Galán et al., arXiv:1910.07398 (2019).
[7] Á. Jiménez-Galán et al., arXiv:2005.10196 (2020).
[8] R.E.F. Silva et al., Nat. Phot. **13** 849 (2019).

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Helicity flip of harmonics from finite and infinite topological nanoribbons

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Graphene has been studied extensively in the past. The most interesting features of this system are its relativistic Dirac points in the band structure. By adding an alternating on-site potential M and a complex next-nearest neighbor hopping t_2, as proposed by F. D. M. Haldane [1], the system, so-called "Haldanite" becomes topologically nontrivial. The alternating on-site potential implies that two different elements constitute the system as, e.g., in hexagonal boron nitride. The complex next-nearest neighbor hopping acts similar to a magnetic field. At certain t_2, a topological phase transition occurs. In the topologically non-trivial phase, such systems show topologically protected edge states. It has been shown that in the simplest, one-dimensional topologically non-trivial systems, the Su-Schrieffer-Heeger chain, the high-harmonic yield strongly depends on whether edge states are absent or present [2-4]. However, in the bulk of a two-dimensional, topological system like Haldanite, the topological phase influences the helicity [5] and the circular dichroism [6] of the emitted light even though edge states do not show up explicitly.
In this work, we consider Haldanite nanoribbons that are made of a single row of hexagons along, say, the x direction. Such a nanoribbon is almost one-dimensional with only two lattice sites in lateral direction y for given x. We show that the nanoribbon emits elliptically polarized light upon illumination by a laser pulse that is linearly polarized along the x direction. A change of the helicity can be observed as well [7] but it is completely different to the one observed for bulk Haldanite. For a fixed next-nearest neighbor hopping t_2, the helicity changes sign at a certain harmonic order, see Fig. 1. This change can be explained by examining the Bloch states of the nanoribbon bulk (where periodic boundary conditions are assumed with respect to the x direction). However, the helicity change at a certain harmonic order is also observed for finite ribbons.

References

[1] F. D. M. Haldane, Phys. Rev. Lett. **61**, 2015 (1988).
[2] D. Bauer and K. K. Hansen, Phys. Rev. Lett. **120**, 177401 (2018).
[3] H. Drüeke and D. Bauer, Phys. Rev. A **99**, 053402 (2019).
[4] C. Jürß and D. Bauer, Phys. Rev. B 99, 195428 (2019).
[5] R. E. F. Silva et al., Nature Potonics **13**, 849 (2019).
[6] A. Chacón et al., arXiv:1807.01616v3 [cond-mat.mes-hall].
[7] C. Jürß and D. Bauer, arXiv:2006.16037 [cond-mat.mes-hall].

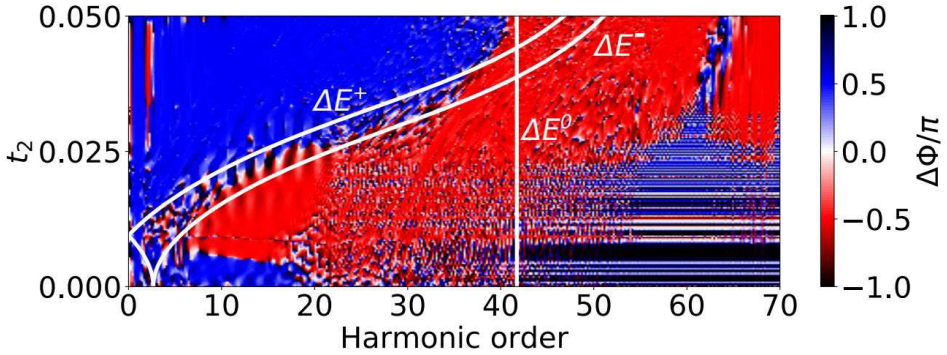

Figure 1. Ellipticity of the emitted light as function of the next-nearest neighbor hopping t_2 and the harmonic order, from [7]. The origin of the sudden change in helicity between the two white lines will be explained in the presentation.

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Cluster approach to attosecond photoelectron emission from dichalcogenides

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We analyze photoemission cross sections and Wigner time delays from WSe_2 for W(4f), Se(3d) and Se(4s) states, following the experiment by Siek et al. in Ref. [1]. We work with progressively larger model clusters to understand the photoelectron diffraction and scattering effects by neighboring atoms, see Fig. 1(a). The calculation scheme is based on a DFT one-center partial-wave expansion supplemented by smaller localized off-center expansions inside spheres placed at each atomic center [2,3]. The radial-coordinate elements of the basis, main- and off-center expansions, are comprised of B-spline sets, and the exchange and correlation is described by the LB94 functional [4]. The approach makes use of group theory to reduce the computational costs; all the systems presented in Fig. 1 belong to the D3h point group, with the polarization vector along the molecular axis, i.e., normal to the surface. Fully differential cross sections show clear scattering effects and indicate deflection of the emitted photoelectron by the neighboring atoms, see Fig. 1(b). We stress that our calculations naturally incorporate all scattering effects and allow us to study the angular dependence of the photoemission cross sections and the Wigner time delays in the attosecond scale.

References

[1] F. Siek et al., Science, **357** 1274-1277 (2017).
[2] D. Toffoli et al., Chemical Physics, **276** 25-43 (2002).
[3] D. Toffoli et al., Journal of Chemical Theory and Computation, **12** 4996-5008 (2016).
[4] R. van Leeuwen et al., Physical Review A, **49** 2421 (1994).

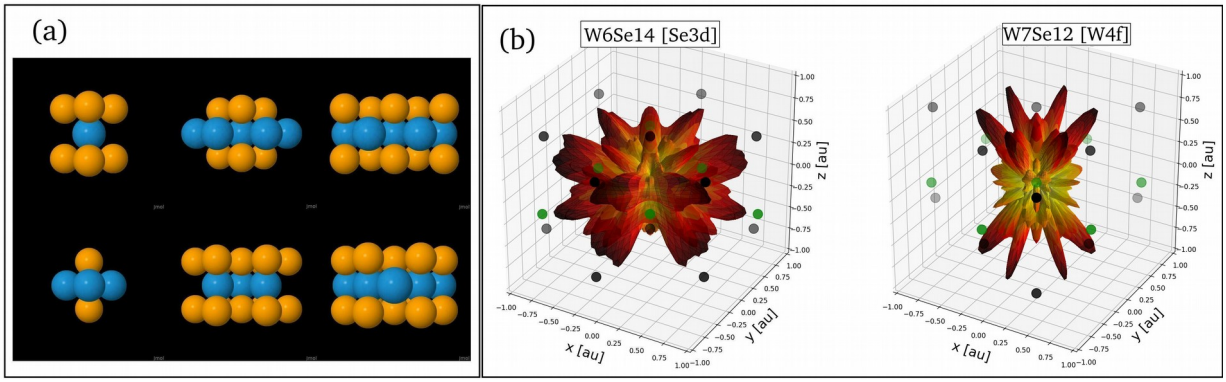

Figure 1. (a) From left to right, top: WSe_6, W_7Se_6, W_7Se_{12}; bottom: W_3Se_2, W_3Se_{14}, W_6Se_{14}. (b) Fully differential cross sections from W_6Se_{14} clusters [Se(3d) orbital] and from W_7Se_{12} [W(4f) orbital].

Attosecond metrology of phase-coherent multi-PHz currents in bulk solids

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A vigorous research activity has followed after first demonstration of high-harmonic-generation from bulk solids[1]. HHG in solids has been demonstrated in THz[2], visible[3] and extreme-ultraviolet spectral ranges[4]. However, underlying mechanism of HHG in solids is deeply debated. A generalized re-collision picture of electron-hole pairs akin to HHG in gases and scattering of carriers in electronic bands (intraband currents) are the two competing mechanisms. We used attosecond streaking to characterize temporal structure of EUV emission from a bulk dielectric medium[5] (SiO_2). The fine details of the dynamic response of the system encoded in the chirp of the emerging attosecond pulse enabled the establishment of a firm link between the emission of EUV radiation and the intraband electric currents induced by the field-driven, coherent, nonlinear scattering of charge carriers in the bulk of SiO_2[5].

To study the phase coherence of induced multi-PHz currents (EUV waveforms), we have employed a novel photoelectron interferometric technique[5,6]. EUV waveforms were found to be reproducible by $\pi/20$ rad in their carrier envelope phase (CEP). We have further studied the dependence of phase of EUV emission on intensity and CEP of the driving laser pulses, and contrasted the results directly with those of EUV emission from a noble gas. Dipole phase in atomic HHG is known to depend linearly on driving pulse intensity and on it's CEP. Our study transparently confirms this; linear phase of EUV waveforms (i.e., their CEP) and nonlinear phase (group delay dispersion) were found to increase with increase of CEP as well as intensity of the optical driver. Contrary to the above findings in gas phase HHG, phase of EUV emission from bulk dielectrics was found to be immune to CEP and intensity variation of the optical driver[6]. Our approach of tracing the phase of HHG via photoelectron interferometry can be well used to decipher mechanisms of HHG.

References

1 Ghimire, S. *et al.* Observation of high-order harmonic generation in a bulk crystal. *Nat. Phys* **7**, 138-141 (2011).

2 Schubert, O. *et al.* Sub-cycle control of terahertz high-harmonic generation by dynamical Bloch oscillations. *Nature Photon.* **8**, 119-123 (2014).

3 Vampa, G. *et al.* Linking high harmonics from gases and solids. *Nature* **522**, 462-464 (2015).

4 Luu, T. T. *et al.* Extreme ultraviolet high-harmonic spectroscopy of solids. *Nature* **521**, 498-502 (2015).

5 Garg, M. *et al.* Multi-petahertz electronic metrology. *Nature* **538**, 359-363 (2016).

6. Garg, M. *et al.* Ultimate waveform reproducibility of extreme-ultraviolet pulses by high-harmonic generation in quartz. *Nat. Photonics* **12**, 291–296(2018)

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Origin of strong-field-induced low-order harmonic generation in amorphous quartz

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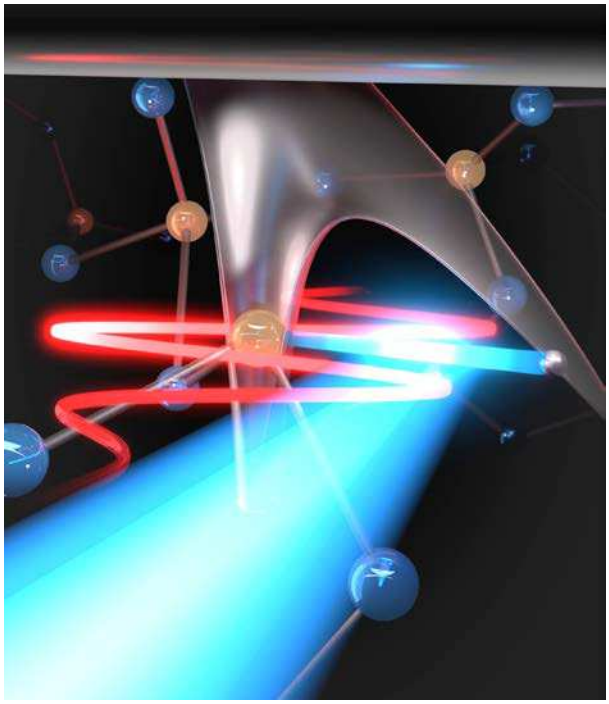
Kerr-type nonlinearities form the basis for our physical understanding of nonlinear optical phenomena in condensed matter, such as self-focusing, solitary waves and wave mixing. In strong fields, they are complemented by higher-order nonlinearities that enable high-harmonic generation, which is currently understood as the interplay of light-driven intraband charge dynamics and interband recombination. Remarkably, the nonlinear response emerging from the subcycle injection dynamics of electrons into the conduction band, i.e. from ionization, has been almost completely overlooked in solids and only partially considered in the gas phase in terms of the so-called Brunel mechanism. Here, we reveal this strong-field-induced nonlinearity in a-SiO2 as a typical wide-bandgap dielectric by means of time-resolved, low-order wave-mixing experiments, and show that, close to the material damage threshold, the so far unexplored injection current provides the leading contribution [1]. Viewed in an atomic picture, this injection current results from the electronic current inside the tunnel – and not from the subsequent field-driven motion as suggested by Brunel.



In this talk I will discuss the underlying experimental results and a model that enables to associate the measured time-resolved low-order harmonic emission with the injection current. The sensitivity of the harmonic emission to the subcycle ionization dynamics may offer an original approach to characterize the evolution of laser-induced plasma formation in optical microprocessing.

References

[1] P. Jürgens, B. Liewehr, B. Kruse, C. Peltz, D. Engel, A. Husakou, T. Witting, M. Ivanov , M. J. J. Vrakking, T. Fennel, A. Mermillod-Blondin, Nature Physics 2020, published online (https://doi.org/10.1038/s41567-020-0943-4)

Attosecond transient absorption spectroscopy in condensed matter systems

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New technologies from the last decade allow for the production of petahertz laser fields, which may be used to drive ultrafast currents in periodic systems. It is then important to design schemes to extract time-resolved information about the electron dynamics in this short time scales.
In attosecond X-ray transient absorption (ATA) spectroscopy an attosecond probe laser pulse is used to promote electrons from core bands to valence/conduction bands during the coherent dynamic of the electrons under an IR petahertz laser pulse. Since core bands are almost flat, all the retrievable information can be linked to the band energy dispersion that the electrons feel during their light-induced trajectories. Furthermore, ATA could also contain information of the intraband Berry phase acquired during the electron propagation. Using as a sample monolayer graphene, we explain how ATAs appear to be a successful tool to obtain measurable properties, both geometrical and dynamical, of the system. Our theory shows good agreement with numerical simulations of a time-dependent density matrix [1, 2].

This is the first theoretical exploration of attosecond X-ray transient absorption spectroscopy in 2D materials, paving the way to the understanding of ATA spectra in 2D and bulk materials. It is expected that the theory used can help learning new features related to coherent light-induced petahertz dynamics in modern material, fundamental for the development of novel quantum devices.

References

[1] A. Picón et al., "Attosecond x-ray transient absorption in condensed-matter: a core-state-resolved Bloch model", New Journal of Physics, **21** 043029 (2019)

[2] B. Buades et al., "Attosecond-resolved petahertz carrier motion in semi-metallic TiS_2", arXiv:1808.06493

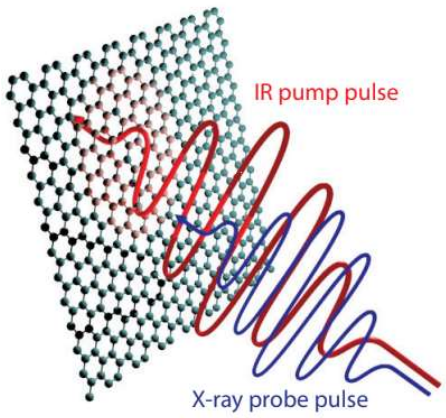

Figure 1 ATA scheme in graphene monolayer

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High order harmonic generation spectroscopy of ZnTe

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High-order harmonic generation (HHG) is the cornerstone of attosecond science. This highly non-linear optical phenomenon has been extensively used during the past few decades for probing electronic structures and dynamics of gaseous media on their natural scale [1]. The shift of the strong laser technology towards higher wavelengths has recently enabled HHG from solid targets [2], which opens up new perspectives for bringing the attosecond technology and spectroscopy tools to the condensed state.

We report on our recent developments in this direction. We developed a three-stage optical parametric amplifier delivering sub-60 fs mid-infrared pulses tunable in the spectral region between 2.7 μm to 4 μm [2] with a maximum energy of 350 μJ. We demonstrate the capability of this source to generate high-order harmonics in semiconductor crystals such as ZnTe and Ge both below and above the bandgap.

We studied the angular dependence of the harmonic yield as a function of the polarization direction of the driving laser field with respect to the crystal axis. The results obtained in a 500-μm thick ZnTe crystal cut along the <110> plane are reported in Fig. 1(a). A four-leaf clover pattern can be observed for all even harmonics, while for odd harmonics an additional lobe along the clover bisectors appears. We can interpret these results in the re-collision framework, where the HHG emission comes from collisions between the excited electron and the nearest neighboring atoms. Within this picture, we can reconstruct the expected pattern for odd (left) and even (right) harmonics, which is reported in Fig. 1(b). The main structure of the harmonic angular dependence is captured by the model.

This outcome demonstrates that HHG in crystals can be exploited as a sensitive probe of the structure and orientation of the crystalline medium. Motivated by these experimental findings, we want to apply more complex HHG spectroscopy techniques to the solid-state. In particular, in our laboratories, we can exploit an additional strong field OPA source tunable around 1.5 μm and a THz source for pump-probe experiments of electron dynamics. The aim is to detect the transient modification of the electronic band-structure after or during the excitation of a second pulse.

References

[1] C. Vozzi et al. Nature Physics, **7** 822-826 (2011)
[2] S. Ghimire et al.Nature Physics, 7 138–141 (2011).

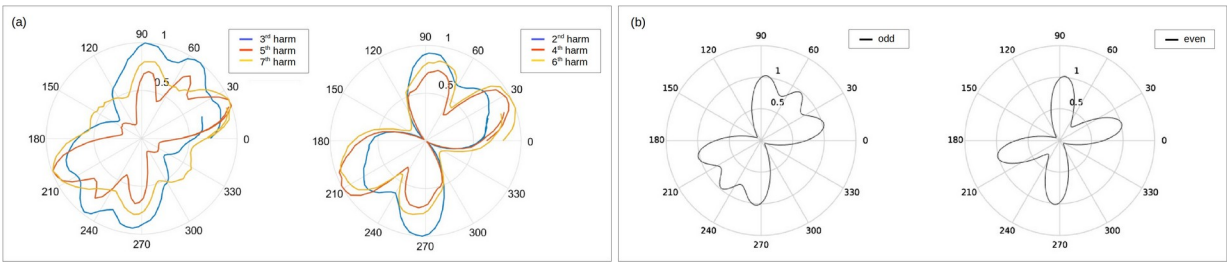

Fig. 1. (a) Experimentally measured orientation dependent yields of harmonics. (b) Simulated orientation dependent spectra of an odd (left) and even (right) harmonic.

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Signatures of electronic Talbot revivals in HHG from a one-dimensional crystal

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High-order harmonic generation (HHG) is considered as a robust technique to generate extreme-ultraviolet and soft x-ray coherent radiation [1], emitted in the form of attosecond pulses. A typical HHG spectrum is characterized by a few low-order harmonics with decreasing energy, followed by a non-perturbative plateau, where the intensity of the harmonics remains approximately constant until a cut-off frequency. HHG is based on tunnel ionization, acceleration and recombination of an electronic wave-packet upon interaction of an intense laser field with an atomic or molecular system. Recently there has been an increasing interest to drive HHG in crystalline solids [2,3]. The characteristic periodicity of these systems leaves a signature in the electronic wave-packet dynamics after ionization. In this work, we introduce a novel phenomenon in HHG, directly related with the coherent wave nature of the ionized electronic wave-packet. The space-time analogy between the propagation of light in the Fresnel approximation and the time evolution of the electronic wave-packet in the Schrödinger equation leads to the prediction of Fresnel diffraction phenomena in the electronic dynamics. In particular, we report the consequences of electronic Talbot effect –analogous to the well-known optical Talbot effect [4]– in the HHG from electrons ionized from periodic structures. Through temporal modulations in the electronic density of probability during its excursion in the continuum, Talbot images lead to the generation of a series of ultra-high frequency harmonics, well above the HHG cut-off frequency.

We numerically solve the two-dimensional time-dependent Schrödinger equation in a one-dimensional crystalline solid with a lattice constant of 2.12 Å, modelled by a Mathieu-type potential. For the states at the extreme of the Brillouin zone, we obtain a spectrum that extends towards the x-ray regime, as shown in figure 1. The dashed lines correspond to the Talbot frequencies, which are calculated from the matter-analogous Fresnel integral [5]. The agreement between the calculated Talbot frequencies and the HHG spectrum confirm the direct relationship of the matter Talbot effect with the generation of these ultra-high-order harmonic frequencies.

We thus demonstrate a novel strategy to control the HHG process from crystalline solids, based in the coherent propagation of the ionized wave-packet and the production of coherent ultra-high frequency radiation up to the x-ray regime. Furthermore, our findings represent a potential spectroscopic method as the HHG reveals spatial features of the electronic wavefunction of the system.

References

[1] F. Krausz et al., Rev. Mod. Phys., **81** 163-234 (2009)
[2] S. Ghimire et al., Nat. Phys., **7** 138-141 (2011)
[3] N. Tancogne-Dejean et al., Sci. Adv., **4**, eaao5207 (2018)
[4] J. Wen et al., Adv. Opt. Photonics, **5**, 83-130 (2013)
[5] C. Granados et al, Phys. Rev. A, **89**, 023428 (2014)

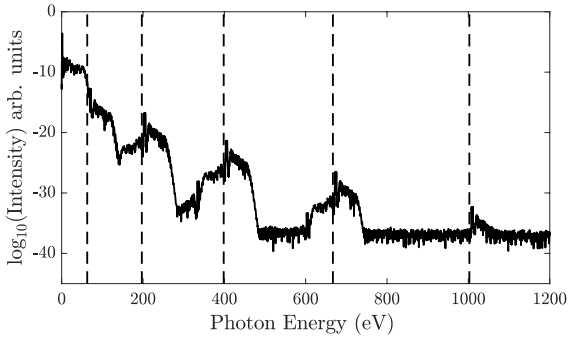

Figure 1. HHG spectrum for the state in the extreme of the Brillouin zone and a driving wavelength of 800 nm. The dashed lines represent the theoretically predicted Talbot frequencies.

Following Phonon Dynamics in Solids Using High-Harmonic Spectroscopy

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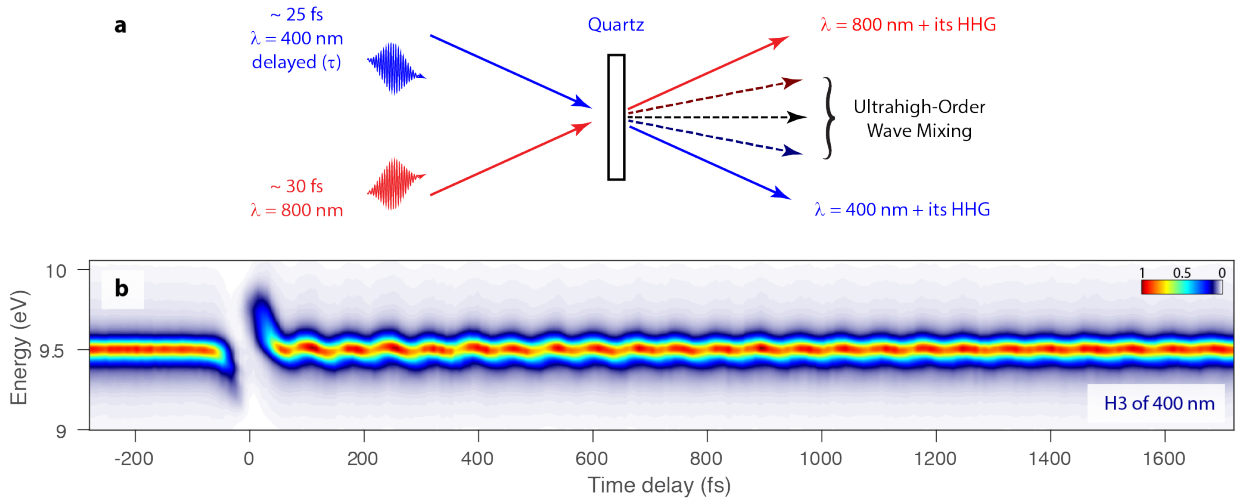

Figure 1: a, Experimental scheme used to perform non-collinear two-color interferometric measurements of HHG from quartz. **b,** Time-resolved spectrum of the third harmonic of 400 nm laser pulses passing through the sample after being excited by the pump 800 nm laser pulses.

Ever since the first observation of high-order harmonic generation (HHG) from zinc-oxide [1], the field of HHG from solids has seen tremendous growth with many reports of HHG from variety of materials as well as their applications in spectroscopy or potential industrial market [2,3,4,5]. HHG from solids has quickly formed a significant sub-field of attosecond science. In this work, we demonstrate the substantial power of high harmonic spectroscopy of solids in investigating carrier-phonon scattering in solids. Utilizing the non-collinear two-color interferometric measurements, we obtain unambiguous ultra-high-order wave mixing in solids, extending from the gas phase experiments [6]. Furthermore, by exercising control over the pump and probe laser pulses as well as their delay and polarization, we can initiate, control, and follow the phonon dynamics in real time with unprecedented capabilities. The experimental results are fully supported by theoretical simulations using different methodologies (two-level model including phenomenological phonon coupling, semiconductor band model including carrier-phonon interaction). Being coupled with theoretical simulations, our experimental results would help in characterizing the carrier-phonon coupling amplitude, phonon creation and relaxation, multiple-phonon dynamics in a solid-state quantum system. We anticipate our work would help revealing carrier-phonon scattering in a brand-new dimension, extracting in-depth information that was not possible otherwise.

References

[1] S. Ghimire *et al.,* Observation of high-order harmonic generation in a bulk crystal, Nat. Phys., **7**, 138-141 (2011)
[2] O. Schubert *et al.,* Sub-cycle control of terahertz high-harmonic generation by dynamical Bloch oscillations, Nat. Photonics, **8,** 119-123 (2014)
[3] M. Garg *et al.,*Multi-petahertz electronic metrology, Nature, **538**, 359-363 (2016)
[4] T. T. Luu, H. J.Wörner, Measurement of the Berry curvature of solids using high-harmonic spectroscopy, Nat. Commun. **9**, 916 (2018)
[5] G. Vampa *et al.,* Plasmon-enhanced high-harmonic generation from silicon, Nat. Phys. **13**, 659 (2017)
[6] J. B. Bertrand *et al.,* Ultrahigh-order wave mixing in noncollinear high harmonic generation, Phys. Rev. Lett. **106**, 023001 (2011)

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Ab initio study of high-order harmonics and isolated attosecond pulses from a solid target

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On the basis of real-time *ab initio* calculations, we study the non-perturbative interaction of strong two-color laser pulses with MgO crystal. The focus of this study is to evaluate the possibility of generating an isolated attosecond pulse (IAP) from high-harmonic emissions from MgO crystal. In this regard, we examine the impact of incident pulse characteristics such as its shape, intensity, and ellipticity as well as the consequence of the crystal anisotropy on the emitted harmonics and their corresponding IAPs. Using infrared asymmetric pulses with a duration of 18 fs and an intensity of ~ 10^{13} W/cm^2, our calculations predict the creation of IAPs with a duration of ~300 attosecond; this is shorter than what was measured experimentally or predicted theoretically in others crystalline systems. It has been shown that the HHG signals drop for the elliptically polarized pulses; however, it provides the availability of easily generate elliptically polarized IAPs. Harmonics cutoff scales linearly with the driving pulse peak field; moreover, the cutoff dependency to pulse wavelength, pulse ellipticity, and the crystal anisotropy are discussed. Our results show the potential of solid-state materials in future ultrafast devices with novel applications beyond the traditional gas systems.

Mechanism for high-order harmonic generation in graphene and carbon nanotubes

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The mechanism for high-order harmonic generation (HHG) in atoms and molecules is well understood in semiclassical terms. According to the three step model [1, 2], the electronic wave packet is promoted into the continuum by tunneling ionization upon the interaction with an intense electromagnetic field; after the release, the wave packet is accelerated by the field and driven back towards the parent ion, to subsequently release the acquired kinetic energy in the form of high-frequency radiation. This simple mechanism explains the observed scaling of the spectral cut-off frequency as $\hbar\omega_c \propto I\lambda^2$. In contrast, the cut-off frequency has been found to scale near to linearly with the field amplitude and then to saturate, in finite-gap solids [3]. In this contribution we present theoretical results of HHG in low dimensional carbon allotropes: 2D single layer graphene (SLG) and 1D single-wall nanotubes (SWNTs), induced by intense few-cycle laser pulses at infrared wavelengths. Our results show the emergence of a non-perturbative spectral plateau at large intensities but, unlike the above referred systems, there is no simple law governing the scaling of the cut-off frequency. Interpreting this particular behavior of the cut-off frequency scaling allows us to unveil the fundamental mechanism for HHG in these low dimensional carbon allotropic structures. Using a saddle-point approximation model (SPAM) for the emission dipole, we show that the first step for HHG, both in SLG and SWNTs, is radically different from the tunneling ionization/excitation process found in atoms, molecules and finite gap materials, and that it is closely related to the topological features of the electronic band structure. In the case of graphene, we show that the high order harmonic radiation is initiated by the non-adiabatic crossing through the Dirac points caused by the singular behavior of the Berry connection [4], while for SWNTs the electron-hole creation is connected to non-adiabatic crossing through the first van Hove singularity of the bands closest to the Fermi level [5].

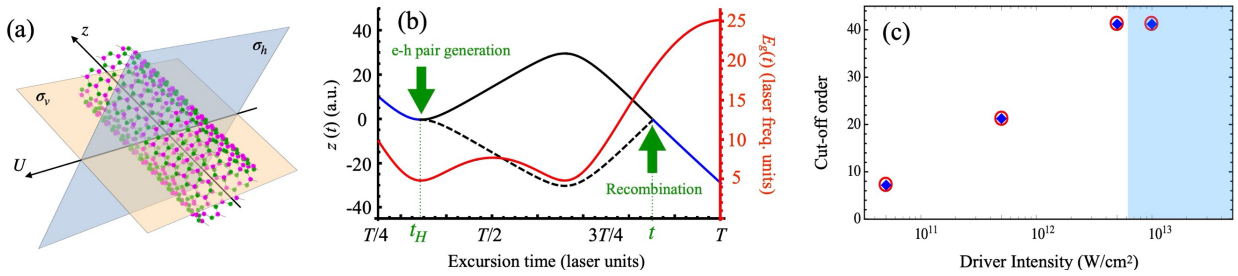

Figure 1. (a) View of the structure and symmetries of the (9,9) armchair nanotube. Magenta and green circles represent the atoms in the two sublattices of the unrolled graphene sheet. (b) Mechanism for HHG in graphene and carbon nanotubes. The e-h pair is produced by the non-adiabatic crossing of Dirac points or van Hove singularities. (c) Spectral cut-off orders as a function of the driver intensity for SWNT, circles and diamonds correspond to our semiclassical model prediction and the numerical integration of the Schrödinger equation.

References

[1] P. B. Corkum, "Plasma perspective on strong field multiphoton ionization", Phys. Rev. Lett. **71**, 1994 (1993).
[2] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, "Above threshold ionization beyond the high harmonic cutoff", Phys. Rev. Lett. **70**, 1599 (1993).
[3] G. Vampa, C. R. McDonald, G. Orlando, D. D. Klug, P. B. Corkum, and T. Brabec, "Theoretical Analysis of High-Harmonic Generation in Solids", Phys. Rev. Lett. **113**, 073901 (2014).
[4] O. Zurrón, A. Picón, and L. Plaja, "Theory of high-order harmonic generation for gapless graphene", New J. Phys. **20**, 053033 (2018).
[5] O. Zurrón-Cifuentes, R. Boyero-García, C. Hernández-García, and L. Plaja, "High harmonic generation in armchair carbon nanotubes", Opt. Express **28**, 19760 (2020).

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A comprehensive study of momentum matrix elements for Bloch states using a local orbital basis

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The linear momentum operator $\hat{\boldsymbol{p}}$ is one of the building blocks of the quantum mechanics formulation. Although its resulting formulas in atomic problems are widely known and are of a basic knowledge in the community, the counterpart formulation in solid state physics presents technical problems not fully appreciated nowadays. From a single-particle standpoint, the discussion started two decades ago on the usability of the formula $\boldsymbol{p}(\boldsymbol{k})=\nabla_{\boldsymbol{k}} H(\boldsymbol{k})$, which only holds in coordinate representation, but that has been misused for variational ab-initio calculations, where $H(\boldsymbol{k})$ becomes an Hermitian matrix. By working with a local orbital basis, Pedersen *et al.* [1] added some correction terms in the previous expression and recently a very general formula was given by Lee *et al.* [2]. Their results relies on the application of the formula $\langle n\boldsymbol{k}|\hat{\boldsymbol{p}}|n'\boldsymbol{k}\rangle = i[\epsilon_n(\boldsymbol{k})-\epsilon_{n'}(\boldsymbol{k})]\langle n\boldsymbol{k}|\hat{\boldsymbol{r}}|n'\boldsymbol{k}\rangle$, which naively follows from the fundamental equation $\hat{\boldsymbol{p}}=i[\hat{H},\hat{\boldsymbol{r}}]$ when projected in a Bloch basis with $n\boldsymbol{k}$ quantum numbers. In parallel, Gu *et al.* [3] worked out another expression using a pure real-space representation, showing that the actual result should be $\langle n\boldsymbol{k}|\hat{\boldsymbol{p}}|n'\boldsymbol{k}\rangle = i[\epsilon_n(\boldsymbol{k})-\epsilon_{n'}(\boldsymbol{k})]\langle n\boldsymbol{k}|\hat{\boldsymbol{r}}|n'\boldsymbol{k}\rangle + \langle n\boldsymbol{k}|\hat{\boldsymbol{C}}|n'\boldsymbol{k}\rangle$, where the additional term is evaluated on the surface of the material. This is in apparent contradiction with Lee *et al.* results where a surface term is absent. Here we take on this issue, trying to clarify the origin of the discrepancy and pointing at the many pitfalls that has been overlooked on recent year treatments when using a local orbital basis. After quantifying the different terms on the equalities, we provide numerical examples to the real effect of the approximations (or unadverted errors) in single-particle standard calculations, as well as the effect of the (necessary) finite size of the Hilbert Space in ab-initio methods.

References

[1] T. G. Pedersen et al., Phys. Rev. B **63**, 201101 (2001).
[2] C-C. Lee et al., Phys. Rev. B **98**, 115115 (2018)
[3] B. Gu et al., Phys. Rev. B **87**, 125301 (2013)

Low order harmonics of an organic polymer

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High-harmonic spectroscopy has developed to a versatile tool to probe structure and ultrafast light-induced processes in complex systems, ranging from multi-electron dynamics in molecules [1] to the electronic band structure of solids [2]. We aim to use harmonic spectroscopy in order to study systems that reside between bulk materials and isolated molecules such as polymers and organic molecular films. The focus of the presented poster is the characterization of the setup for harmonic generation with laser pulses in the mid-infrared. First proof of principle measurements with the organic polymer poly (methyl methacrylate) are presented up to the damage threshold of the material. For this first test target, we find low order harmonics up to order 7 with an intensity scaling of I^n.

References

[1] O. Smirnova et al., Nature **460**, 972 (2009)
[2] G. Vampa et al., Phys. Rev. Lett. **115**, 193603 (2015)

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Beyond-Fröhlich theory of large polarons in anharmonic solids

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In solids where the interaction between electrons and phonons is large, the electrons can be replaced with a quasiparticle to account for this coupling. This quasiparticle is known as the polaron. Fröhlich [1] first derived a Hamiltonian describing so-called 'large' polarons that couple to longitudinal optical phonons in a cubic crystal. This Hamiltonian can be seen as the simplest model for large polarons, and is still used as the basis for analytical investigations today.
In his original derivation, Fröhlich considered a lattice where the phonons are harmonic and do not interact. This approximation is valid in a wide range of materials, but excludes any possible anharmonic effects. Based on earlier work by Kussow [2], we have derived three additional terms in the Hamiltonian that take into account the anharmonicity of the lattice up to third order. These terms describe 3-phonon processes, 2-phonon-1-electron processes, and 1-phonon-2-electron processes respectively. In the spirit of keeping a simple model, we restrict ourselves to the assumptions of the Fröhlich Hamiltonian mentioned above.
We show that all the third order anharmonic terms are zero in crystals with an inversion center: in this case, one needs to look at the fourth order terms. Using perturbation theory, we show that the anharmonic terms can lead to a significant increase in the binding energy and effective mass of one polaron in the weak-coupling limit.

References

[1] H. Fröhlich, Electrons in lattice fields. *Advances in Physics*, **3**, 11, 325-361 (1954).
[2] A.-G. Kussow, Large polaron in an anharmonic crystal lattice. *Int. J. of Mod. Phys. B*, **23**, 1, 19-38 (2009).

Effects of tunnel ionization on non-linear response of solids

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The notion of nonlinear optical phenomena in dielectric solids has been successfully extended to the strong field ionization regime by linking high-order harmonic generation (HHG) to Bloch oscillations and interband recombination [1,2]. Most recently, however, it was shown that these mechanisms cannot explain the emission of low harmonic orders which, instead, are generated by the strong field tunneling excitation dynamics that drive Brunel and injection currents [3]. While the tunneling injection current has been identified as the dominant mechanism, it is so far not known to which extent information about the transient tunneling barrier is imprinted on emitted low-order harmonics.

Here we investigate the injection induced nonlinear optical response, for different tunneling scenarios using a semi-classical, one-dimensional WKB ansatz to drive an ionization-radiation model [4],

$$\dot{\mathbf{J}} = q_e n_0 \left[\frac{q_e}{m_e} \mathbf{E}\rho + \mathbf{v}_0 \dot{\rho} + \frac{\partial}{\partial t}(\mathbf{x}_0 \dot{\rho}) \right]$$

where the first and second term correspond to Brunel acceleration [5] and birth velocity of electrons in the conduction band, respectively, while the third term describes the injection current associated with strong-field induced interband excitation. We characterize the predicted response in terms of an effective order of nonlinearity by employing a two-color wave-mixing scheme [3] and discuss its scaling dependence on the electronic structure and the local field strength.

References

[1] Luu, T. T. et al. Nature **521**, 498 (2015).
[2] G. Vampa, et al. Nature **522**, 462 (2015).
[3] P. Jürgens, et al. Nat. Phys. (2020). https://doi.org/10.1038/s41567-020-0943-4
[4] M. Geissler, et al. Phys. Rev. Lett. **83**, 2930 (1999).
[5] F. Brunel, J. Opt. Soc. Am. B **4**, 521 (1990).

Theory of high-harmonic generation in topological insulators

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In previous work, the influence of topological effects on high-harmonic generation in finite chains was studied [1]. A many-order-of-magnitude enhancement in the high-harmonic generation efficiency has been shown between different topological phases. Shortly thereafter it was discovered that the same effects are observable using the tight-binding approximation [2]. In this work we investigate the bulk of similar systems. This enables us to employ the Bloch theorem to find solutions for the Schrödinger equation.
We present a general theory, similar to the Lewenstein model in atoms [3], for the generation of high-harmonics in the bulk of solids. Many topological insulators, the main focus of this work, consist of two atoms per unit cell. In tight-binding approximation, such solids can be effectively described by 2x2 Bloch-Hamiltonians, leading to two bands. The theory takes topological effects of such systems into account. Exemplary, the theory is applied to the Su-Schrieffer-Heeger chain [4] and the Haldane model [5] in strong laser fields.

References

[1] D. Bauer and K. K. Hansen, Phys. Rev. Lett. **120**, 177401 (2018).
[2] C. Jürß and D. Bauer, Phys. Rev. B **99**, 195428 (2019).
[3] M. Lewenstein et al., Phys. Rev. A **49**, 2117 (1994).
[4] W. P. Su, J. R. Schrieffer, and A. J. Heeger, Phys. Rev. Lett. **42**, 1698 (1979).
[5] F. D. M. Haldane, Phys. Rev. Lett. **61**, 2015 (1988).

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High-energy mid-IR strong-field source for high-order harmonic generation in solids

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Since the pioneering work from Ghimire et al. [1], high order harmonics generation (HHG) in solids has rapidly emerged as the new frontier for probing electron dynamics in the condensed phase in their natural time scales. This has been possible also thanks to the recent advancements of the strong laser field technology in the mid-IR and THz energy range.
We present here a 1-kHz broadband tunable mid-IR three-stage optical parametric amplifier (OPA) for high-order harmonic generation in solids [2]. The OPA is based on Potassium Titanyl Arsenate (KTA) crystals driven by a commercial 25-fs Ti:Sapphire laser. The mid-IR seed pulse is generated in the first stage via intra-pulse difference frequency generation and subsequently amplified in two stages. The output pulses have sub-60 fs duration and the maximum energy of 300 μJ at 3.25 μm, and they are tunable in the range 2.8-3.8 μm (see Figure 1a).

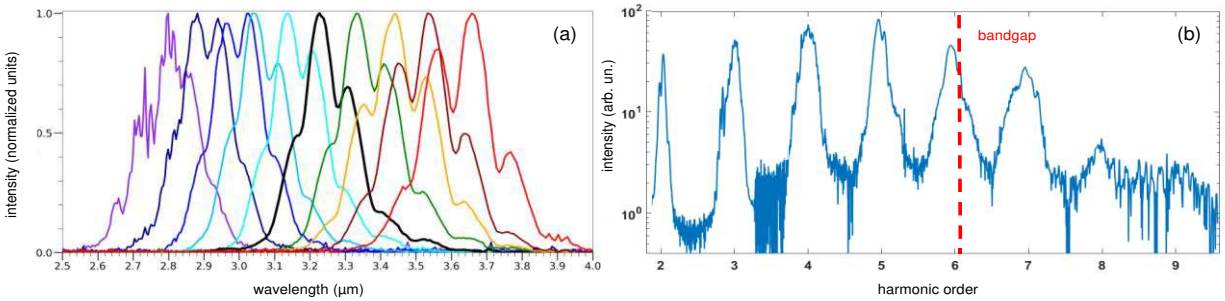

Figure 1(a) Spectra of the mid-IR OPA for different configurations. (b) HHG in ZnTe driven by the OPA tuned at 3.4 μm.

The mid-IR source was exploited for generating higher-order harmonics in semiconductor crystals. We demonstrate the high-order harmonic generation of both even and odd harmonics in ZnTe up to the 8th harmonic order (see Figure 1b). The harmonic emission was studied also as a function of the direction of the crystal axis with respect to the polarization of the driving laser field, demonstrating the dependence of the emission on the symmetry of the crystal (not shown). We also report on HHG in Germanium semiconductor crystals. In this case, only odd harmonics up to the 9th harmonic order have been observed. In both cases, the experimental results demonstrate the capability of this source to generate harmonics in the condensed phase both below and above the bandgap of the semiconductor.
Motivated by these experimental findings, we plan to investigate the role of intraband and interband electronic currents during the high-order harmonic generation process, and to extend and investigate HHG emission from more complex and engineered materials.

References

[1] Ghimire, S. et al., "Observation of high-order harmonic generation in a bulk crystal.", Nature Phys. 7, 138–141 (2011).
[2] Musheghyan, M., et al. "Tunable, few-cycle, CEP-stable Mid-IR Optical parametric amplifier for strong field applications", J. Phys. B (2020) accepted for publication.

Frequency Upconversion of Quadruple-Gaussian Laser Beams by Excitation of Overtones in InSb with Linearly Increasing Doping Profile

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Theoretical study on frequency upconversion of Quadruple-Gaussian (Q.G) laser beams by excitation of frequency overtones on n-type InSb has been presented. Due to narrow band gap structure of InSb the motion of conduction electrons becomes relativistic at moderate laser intensities. As a result the effective mass of the carriers becomes a function of laser intensity via relativistic mass correction. As the natural frequency of oscillations of electrons and hence index of refraction of semiconductors depends on effective mass of the electron, the index of refraction becomes a function of laser intensity. Under the effect of this nonlinear index of refraction the laser beam gets self focused. As a result the oscillation amplitude of the conduction electrons in the field of laser beam becomes very high. The oscillations of conduction electrons contain overtone frequencies of the pump beam and thus oscillating electrons results in emission of harmonic radiations those are overtones of the incident beam. Following moment theory based on hamiltonian formulation equation of motion of beam width has been obtained. Also following hydrodynamical fluid model the power of excited laser overtones has been calculated.