Our goal:
• Use an all optical technique to investigate tunneling dynamics; map attosecond dynamics onto a femtosecond time scale
• Move away from photoelectron spectroscopy– enable experiments on bulk
Outline

• How to read out tunneling ionization with an optical pulse?

• Observation of recollision-free (Brunel) harmonics: from noble gas from bulk transparent solids

• Brunel mixing with two-color fields

• Future: use 1.6 µm IR CEP OPA for bulk
Ionization Regimes

Multi-photon (MPI) plasma-induced spectral blue-shift

Tunnel (TI) harmonics generation

\[
\gamma = \frac{\omega_L \sqrt{2mW_b}}{eE_0}
\]
- Keldysh parameter

\(\gamma > 1\) – tunneling rate slower than laser period

Multiphoton ionization

\(\gamma < 1\) – tunneling rate faster than laser period

Tunnel ionization

Keldysh (1965)
Motivation

• Real-time observation of tunneling ionization dynamics

  – Attosecond angular streaking using circular polarized light and COLTRIMS

We set out to develop an optical read-out technique that can work with bulk solids!
Mechanisms of Higher-Order Harmonic Generation

Highest orders (Corkum): Tunnel ionization:
- \( \chi^{<3>} \), \( \chi^{<5>} \), \( \chi^{<7>} \), …

Brunel: twice-per-cycle tunnel ionization \( \Rightarrow \) step-wise plasma concentration increase \( \Rightarrow \) transverse plasma current \( J_\perp = eN_e v \)

F. Brunel, JOSA B 7, 521 (1990)

Harmonics signal is independent of the final state of electrons!

Logarithm of intensity vs. time for different harmonics orders.

\( \chi^{<3>} E^3 \), \( \chi^{<5>} E^5 \), \( \chi^{<7>} E^7 \)

Plasma wave oscillations

Recollision HHG

\( \chi^{<3>} \), \( \chi^{<5>} \), \( \chi^{<7>} \), …

\( \log(I_q) \)

\( \chi^{<3>} \), \( \chi^{<5>} \), \( \chi^{<7>} \), …

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Previous Work

Classical models for harmonic generation based on high-frequency variation of the tunnel ionization current:

**Theory:**

F. Brunel, *JOSA B* 7, 521 (1990)

These models predict the right magnitude of 3rd and 5th harmonics, but no plateau:


**Experiment:**

“Brunel mixing” in gas.
C.W. Siders *et al.*, *PRL* 87, 263002 (2001)
Driven by laser $\omega_1$, $n_e$ oscillates at $2\omega_1$
$\omega_3=2\omega_1 \pm \omega_2 \rightarrow$ for $\omega_1=\omega_2$ predicts THG and THz emission on the leading pulse edge. (Similar to THz emission via 4 wave mixing $2\omega-\omega-\omega$
Time Dependent Refractive Index Modulation

Analytical expression for nonadiabatic tunnel ionization:

3-D TDSE in good agreement with the Yudin-Ivanov formalism:

Information

Refractive index change:

\[ \Delta n_p \approx -\frac{\omega_p^2}{2\omega^2} \]

\[ \omega_p^2 \propto n_e(t) \]

Time-dependent phase shift:

\[ \Delta \varphi_p(t) \propto \frac{1}{\omega^2} \]

Can be read out only by an optical field

Not accessible in the XUV!
Tunneling Dynamics in Bulk Solids

Proof of principle experiment in gas phase

Strategic goal: To develop a technique for investigation of TI dynamics in bulk solids

Ionization is the starting point for all strong field phenomena…

It is also starting point for optical breakdown

Photo-electrons cannot be observed from bulk material!
Stepwise vs. Smooth Refractive Index Modulation?

\[ \gamma = \frac{\omega_L \sqrt{2mW_b}}{eE_0} \] - Keldysh parameter

Attosecond time structure due to strong dependence of ionization probability on the field strength

TI \quad \gamma \quad MPI

Smooth ramp due to MPI (follows intensity envelope)
Quasi-Linear MPI Ramp

**i(onization)-Spider**
Plasma-Blue-Shift spectral shear interferometry for characterization of ultimately short optical pulses

Attosecond Phase Mask

Time domain:
Attosecond phase mask
Showing tunnel ionization dynamics

Frequency domain:
Harmonic spectra

Advantage:
Brunel harmonics do not depend on the final state of electrons

Harmonic frequencies
\[ N \times 2\omega_{\text{pump}} + \omega_{\text{probe}}, \, N=1,2,3\ldots \]

Question:
How badly is the attosecond time-domain phase mask distorted by pulse propagation?
Spectral Response to Temporal Phase Modulation

Formation of time-dependent attosecond phase mask

Formation of harmonic spectrum
“Spectral scattering”

Ionization loss, mm$^{-1}$

$I(t)$

Time, fs

$n_e/n_0$

Spectral intensity, arb. units

$\omega/\omega_0$

$I_{\text{pump}} = 1.5 \times 10^{14}$ W/cm$^2$

$I_{\text{pump}} = 5 \times 10^{13}$ W/cm$^2$

$I_{\text{pump}} = 3 \times 10^{13}$ W/cm$^2$
Interpreting Spectral Signatures

Model:
Phase mask $\Phi(t)$ with different finite rise time $\theta$

This work: A. Zheltikov and E. Serebryannikov, 3-D propagation code for Brunel and Kerr harmonics based on Yudin-Ivanov formalism

Phase mask for $T/\theta = 9$

Spectra of the resulting laser field for different phase masks

Power ratio between adjacent harmonics orders depends on the speed of electron density release ($\Delta n$ step sharpness)
• Pump: 5 fs, 200 μ J; Probe: ~20 fs, 2 μ J.
• The harmonics are detected in the direction of the weak, cross polarized chirped probe pulse. The pump beam is blocked before the entrance slit of the spectrometer.
• $\omega_{\text{probe}}$ may differ from $\omega_{\text{pump}}$ to see the effect of the phase mask.

$\lambda_c = 750$ nm
Results

Harmonics

experiment

signal detected in the direction of a weak cross polarized chirped probe pulse
Experiment vs. Simulation

**Experiment**

- **H3**
- **H5**
- **H7**

**Simulation**

- 300 mBar Kr
Cross-Correlations

Temporal marginal:
H3, H5, H7 maps integrated over spectrum.

All harmonics follow the same time structure.
Linear Dependence on Probe Intensity

3rd Harmonic, 1mm Argon target

Experiment

Theory

Delay (fs)

Wavelength (nm)

Signal (arb. un.)

Wavelength (nm)

Probe Energy ($\mu$J)

Proof of separation of $\chi^{(3)}$ from Brunel-harmonics:

**Linear intensity dependence of H3 on probe intensity!**

THG spectra measured with pump on are blue-shifted!
Propagation Effects

Spatio-temporal profile of the probe pulse

Probe @780 nm

Filtered THG part

780 nm 5 fs 0.2 mJ

f=40 cm

krypton jet,
P=300 mbar.
Distortion by Propagation

Ar, 0.03 bar
\[ \frac{\partial}{\partial t} \left( \frac{n_e}{n_o} \right), \text{fs}^{-1} \]

Ar, 0.3 bar
\[ \frac{\partial}{\partial t} \left( \frac{n_e}{n_o} \right), \text{fs}^{-1} \]
Interaction Length vs. Pressure

Ar density

Electron density buildup, fs$^{-1}$

Time marginal

Interaction length drops with pressure increase
Chirp of Probe Pulse

3rd Harmonic, 1mm Argon target

The spectral marginal does not depend on the chirp of the probe pulse.
Coherent Control with Pump Pulse

3rd Harmonic, 1mm Argon target

$\int d\omega \Rightarrow \text{Cross-correlation}$

$\int d\tau \downarrow \downarrow \text{Spectrum}$
Chirp of Pump Pulse

3rd Harmonic, 1mm Argon target

Each marginal is normalized to its own maxima

The signal decreases fast with increasing pump chirp
Investigation of TI in bulk solids

Experiment in bulk:
Brunel type harmonic in glass

For measuring several harmonics:
Use an OPA system at 1.5 μm
Are We There Yet?

Linear Polarized Pump

Circular Polarized Pump

Low Probe Power/Intensity

High Probe Power/Intensity
direct probe THG (background)

Target: fused silica

Q: Why is there a signal with circular polarization?
Two Color Experiment

\[ \omega_{\text{pump}} \neq \omega_{\text{probe}} \]
Looking for Beat Modes

THG, probe chirped with 3 mm of FS

THG, probe chirped with 6 mm of FS

\[ N \in \text{integer} \]
Ionization in Quartz

Red curves: with avalanche ionization
Dotted blue: without

$$\gamma_{\text{min}} = 0.6$$

Introducing TW/cm$^2$

$$\tau/\tau_p$$

$$\rho_c$$

$$16 \text{ fs}$$
$$10 \text{ fs}$$
$$50 \text{ fs}$$

$$10^{10}$$
$$10^{11}$$
$$10^{12}$$
$$10^{13}$$
$$10^{14}$$
$$10^{15}$$
$$10^{16}$$
$$10^{17}$$
$$10^{18}$$
$$10^{19}$$
$$10^{20}$$
$$10^{21}$$
$$10^{22}$$
$$10^{23}$$

$$\rho$$

$$\sigma$$

$$|\mathbf{E}|$$

$$U_i$$

$$\tau_r$$

$$\partial \rho / \partial t = W_{PI} (|\mathbf{E}|) + \sigma \rho |\mathbf{E}|^2 - \rho / \tau_r$$
CEP stable 20-Hz IR OPCPA

12.5 mJ FWHM~100 nm @ 1.5 µm

Nd:YAG pump

OPA 1+2

stretcher

DAZZLER

3rd KTP

4th KTP

compressor
to filamentation

O.D. Mücke, A. Alisauskas, D. Sidorov, A. Pugzlys, A. Verhoef

CEP drift
2\textsuperscript{nd} OPA Stage (10 kHz)

non-collinearity angle:
walk-off angle of \(~2^\circ\)

\[ M^2 = 1.13 \pm 0.04 \quad (2\textsuperscript{nd}-stage signal) \]
\[ M^2 < 1.2 \quad (\text{Yb:KGW pump}) \]
IR OPCPA (20 Hz)

Pulses after the 4th stage
4-Fold Self-Compression of mJ IR Pulses

Pulses after the 4\textsuperscript{th} stage

Self-compressed 1.5-µm pulses: >1.5 mJ, 3 optical cycles

Gas: Ar, 5 bar,
Cell length: 140 cm
Filament length: 12-15 cm

Optimal compression:
\( E_{\text{in}}: 2.2 \text{ mJ}, \ E_{\text{out}} = 1.5 \text{ mJ} \)
Throughput: 66\%
Summary

• First direct experimental observation of Brunel harmonics in gas and bulk.

• Attosecond ionization dynamics can be mapped onto a spectral response that is free of recollision contribution.

• Attosecond phase mask is not intuitive but quite robust.

• It is feasible to develop an optical technique instead of registering photo-ionization fragments ⇒ attoscience in bulk.

• Future experiments on bulk: use the CEP 1.5(signal)/3.5(idler) µm OPCPA.

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