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Investigating Resonant Two-Color Photo-Ionization Processes in Atoms and Molecules

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Introduction

We present an experimental tabletop set-up dedicated to investigations of ultrafast processes in atoms and molecules combining near infrared (NIR) and extreme ultraviolet (EUV) radiation pulses. Our experiments will focus on the study of electron correlations in highly excited auto-ionizing resonances by different pump-probe techniques aiming to obtain novel insights into atomic and molecular dynamics.

Methods

- Coherent Legend Duo laser system: 3kHz, 3mJ/pulse, 800nm, 30fs
- Pump-probe setup: 800nm + high harmonic generation (HHG)
- Electron and ion time of flight (eTOF/iTOF) spectrometer: T/ΔT > 100, m/Δm > 300 [1]
- Delay-line velocity map imaging (VMI) with nanosecond time resolution [2]
- Coincidence measurements with eTOF, iTOF and VMI
- Molecular jet for precise gas injection

Setup

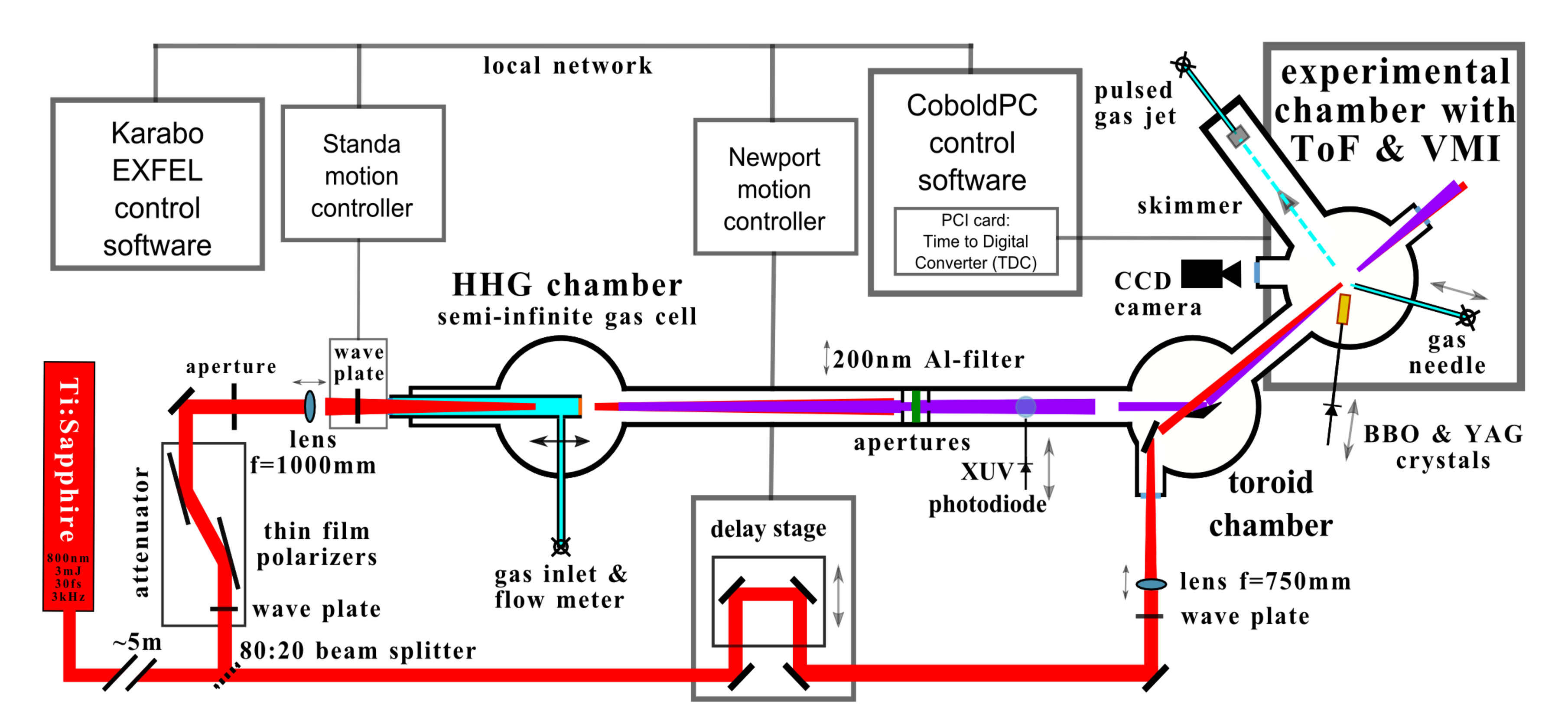

Fig. 1: Schematic drawing of the experimental setup.

HHG in a Semi-Infinite Gas Cell

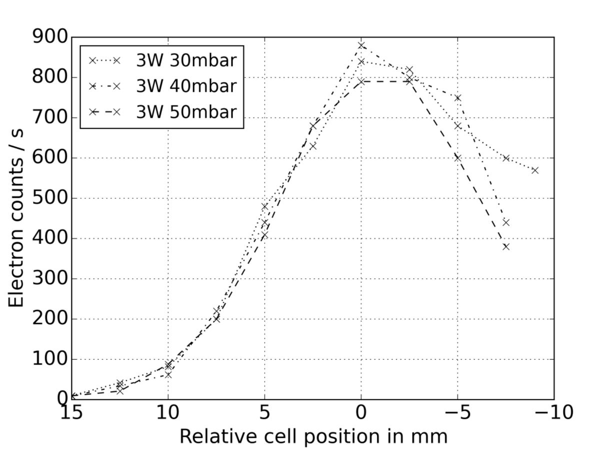

Fig. 2: Influence of pressure and cell position on electron counts released by HHG.

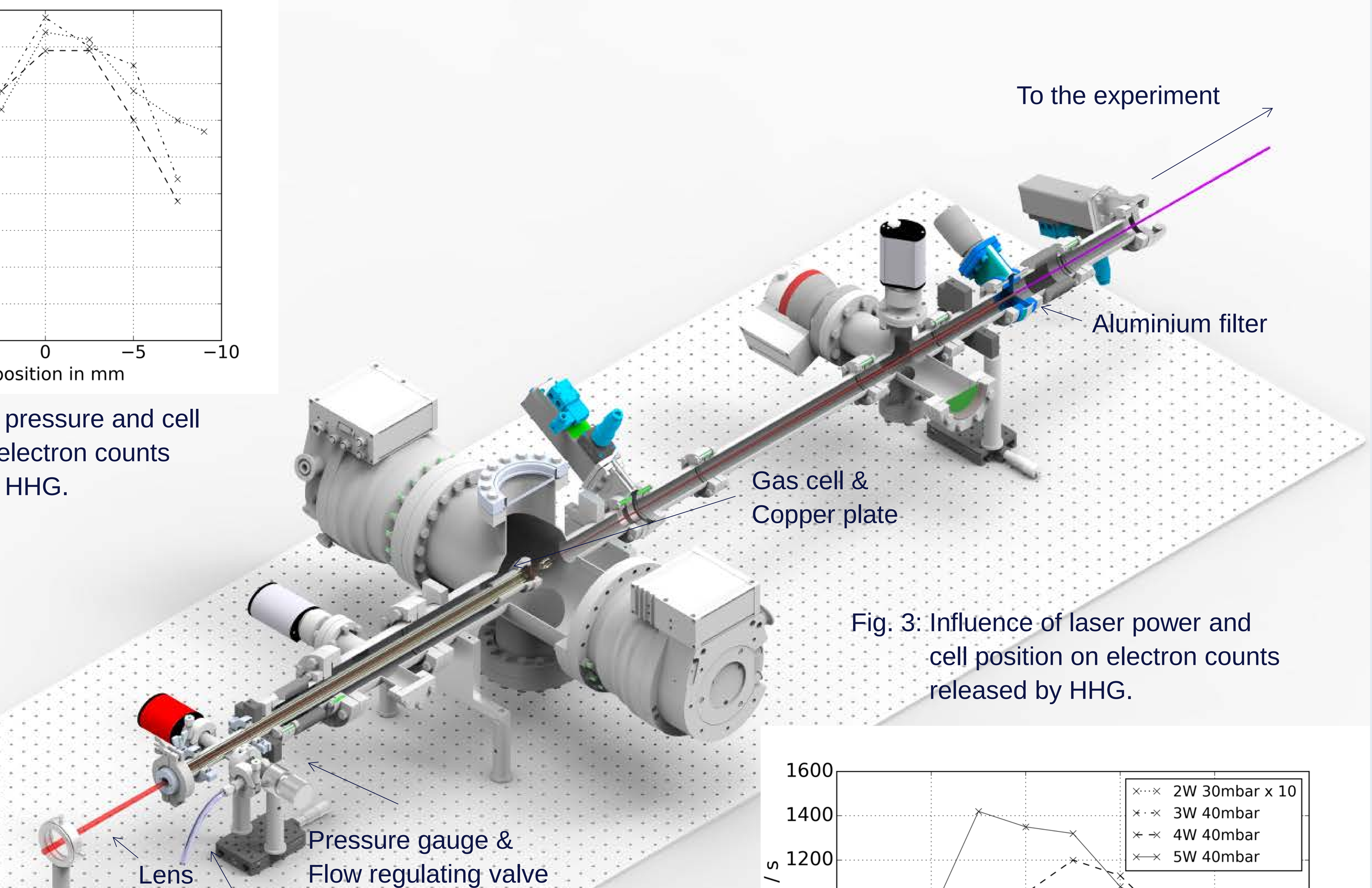

Fig. 3: Influence of laser power and cell position on electron counts released by HHG.

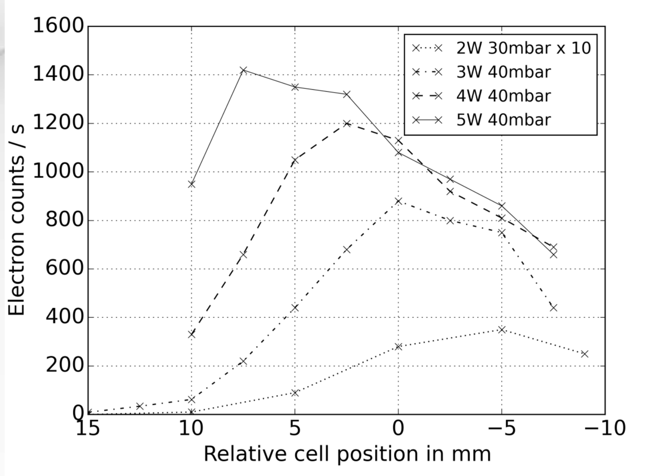

Fig. 4: Setup of the semi-infinite gas cell for the generation of high harmonic radiation.

Phase-matching of HHG

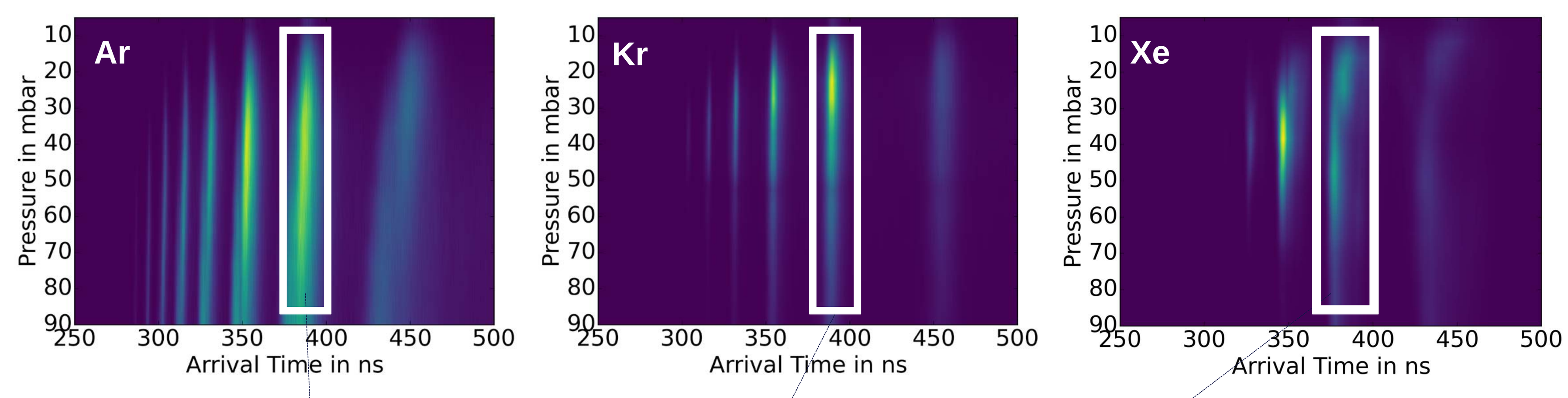

Fig. 5: eTOF spectra of Argon depending on the pressure of the corresponding gas in the HHG cell.

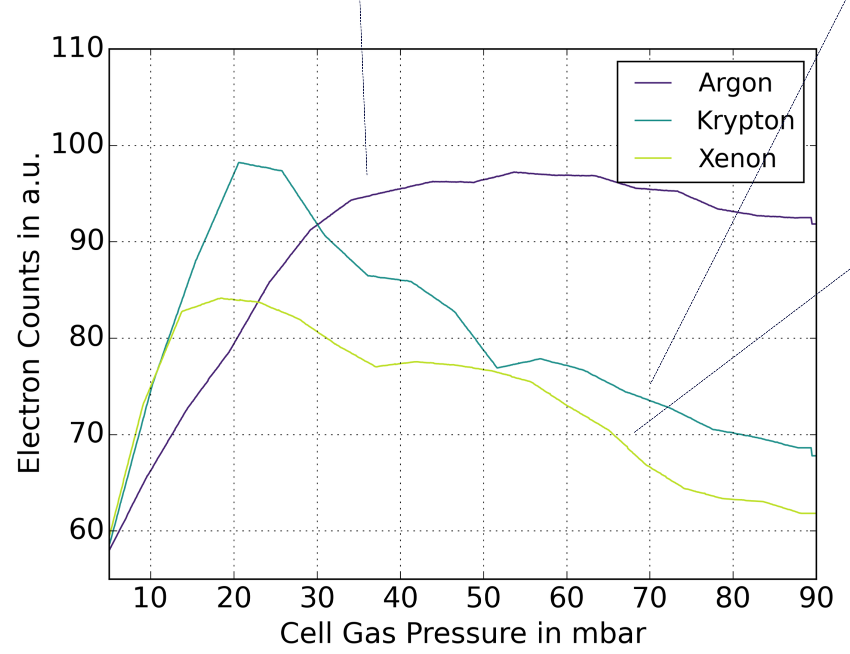

Fig. 6: Electron counts of one high harmonic versus the cell pressure. Compared are three different gases.

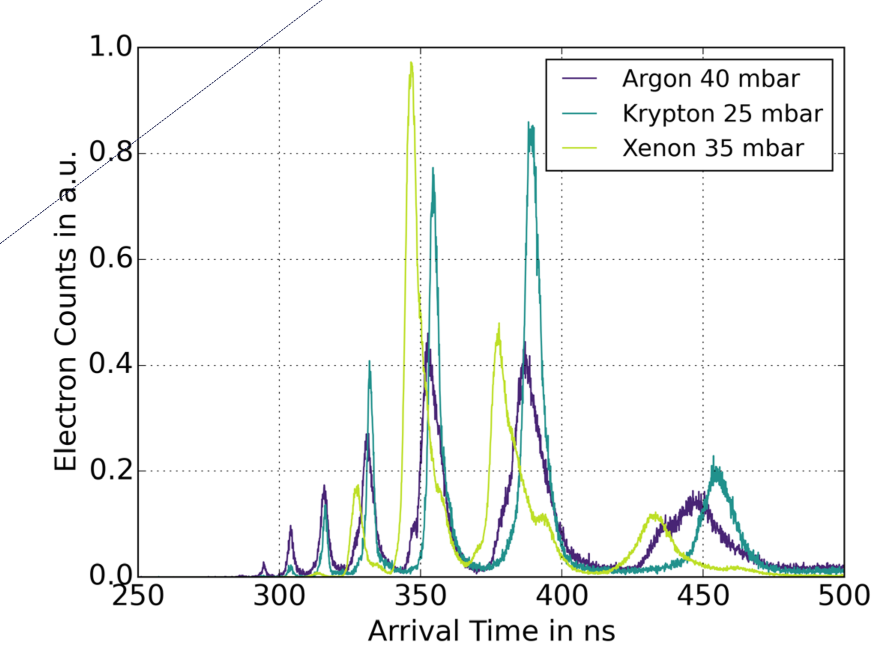

Fig. 7: HHG spectra at optimal pressure for the three gases. Fewer harmonics are supported when heavier noble gases are used to generate HHG

Phase-matching near the focal point depends on the pressure of the neutral gas, the pressure of the plasma and the Gouy-Phase. [3]

$$\Delta k = \Delta k_N(P) + \Delta k_P(P) + \Delta k_G$$

Pulse Characterization with Above Threshold Ionization

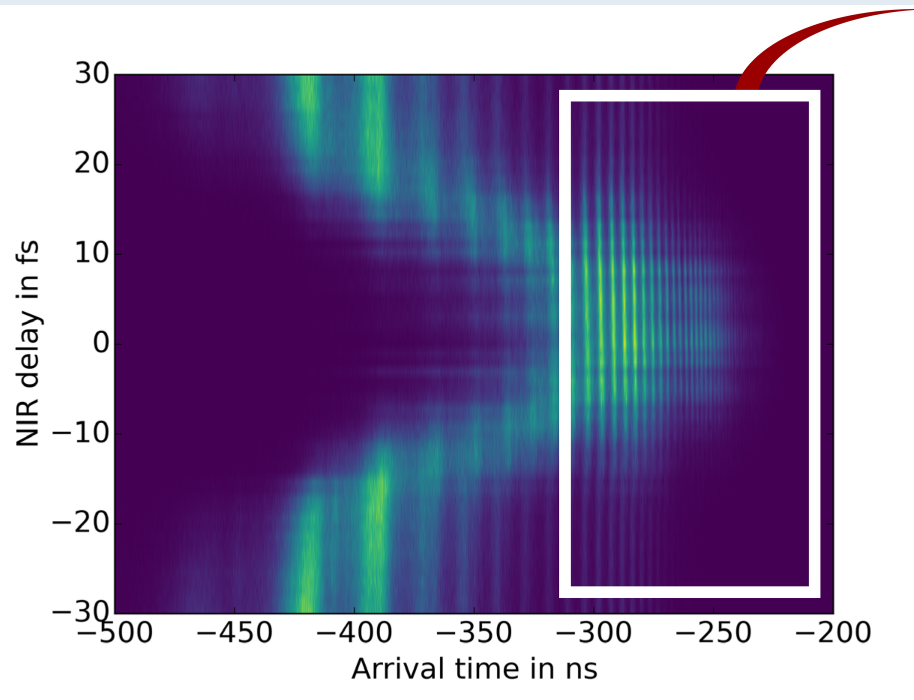

Fig. 8: NIR autocorrelation with above threshold ionization enhancement in Argon

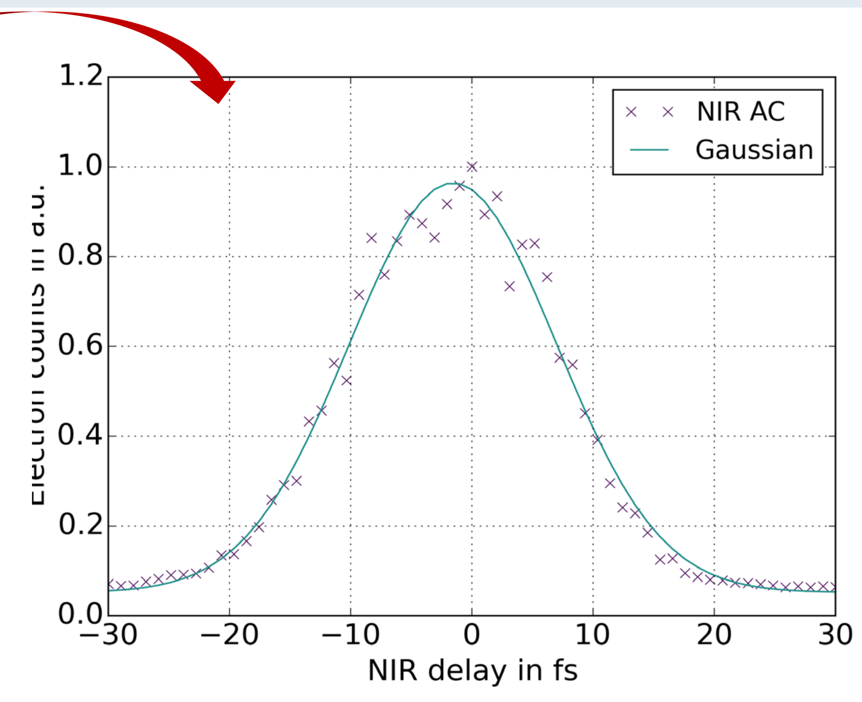

Fig. 9: Integrated NIR autocorrelation over the white box in Fig. 8 Assuming a gaussian beam the pulses FWHM is 28 fs. This result is in line with FROG measurements.

Calculating the temporal width of a laser pulse, assuming the shape is Gaussian:

$$f(x) = a \cdot e^{-\frac{(x-x_0)^2}{2\sigma^2}} + h \quad \rightarrow FWHM = 1.41 \cdot 2.35 \cdot \sigma$$

Outlook

- Calibration
- Second and third harmonic generation
- Commissioning of the molecular jet
- Circular HHG
- Pump-Probe experiments
- Commissioning of the VMI

[1] http://www.kaesdorf.de/ElectronTOF.html (accessed 01/26/2018)
[2] O. Jagutzki, V. Mergel, K. Ullmann-Pfleger, L. Spielberger, U. Spillmann, R. Dörner, H. Schmidt-Böcking, "A broad-application microchannel-plate detector system for advanced particle or photon detection tasks: large area imaging, precise multi-hit timing information and high detection rate", Nucl. Instrum. Meth. A, **477**, 244 (2002).
[3] A. L. Lytle, "Phase Matching and Coherence of High-Order Harmonic Generation in Hollow Waveguides", PhD Thesis, University of Colorado

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