

# Documentation

## Dataset for Manuscript "High-Order Harmonic Generation in an Organic Molecular Crystal"

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## 1 General Information

Dataset title: Dataset for Manuscript "High-Order Harmonic Generation in an Organic Molecular Crystal"

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## 2 Description

This document provides the data, which is discussed in the publication [1].

### 3 File Format

The data is provided in the form of ".txt"-files containing the research data in tables. It can be easily accessed in python with the numpy library using the "numpy.loadtxt( "filename.txt")" command or in matlab with "readtable( "filename.txt")". All entries use double-precision floats and row-major ordering.

### 4 Repository Structure

The data is grouped into the following sub-folders and files:

```
HHG_OMC_Repository
├── Absorption_Spectra
│   ├── OD.txt
│   ├── Polarization.txt
│   └── Wavelength.txt
├── Polarization_Dependent_Yields
│   ├── Yield_Experiment.txt
│   ├── Yield_MR_CIS.txt
│   ├── Yield_TB.txt
│   ├── Yield_TB_flipped_t2_0_01.txt
│   └── Yield_TB_flipped_t2_0_0001.txt
├── Tight_Binding_Flipping_Analysis
│   ├── phi_of_maximum_yield.txt
│   └── t2_values.txt
├── Single_HHG_Spectra
│   ├── Spectrum_Experiment.txt
│   ├── Spectrum_MR_CIS.txt
│   └── Spectrum_TD_DFT.txt
└── Documentation.pdf
```

#### 4.1 Absorption\_Spectra

Includes the data supporting Extended Data Figure 1.

OD.txt	Optical density of the measured polarizations and wavelengths
Polarization.txt	Polarization of the driving field in [°]
Wavelength.txt	Wavelength in [nm]

#### 4.2 Polarization\_Dependent\_Yields

Includes the data supporting all shown polar plots, i.e. Fig. 2, 3 and Extended Data Figure 3. In all data files we provide the harmonic yield and the corresponding laser polarization.

Yield_Experiment.txt	The first row contains the laser polarization, and the following rows contain the experimentally measured harmonic yield of harmonic order 3 to 15 (Fig. 2, 3)
Yield_MR_CIS.txt	The first row contains the laser polarization, and the following rows contain the harmonic yields of order 1 to 13 for the combined emission of two uncoupled molecules calculated with MR-CIS (Fig. 3)
Yield_TD_DFT.txt	The first row contains the laser polarization, and the following rows contain the harmonic yield of harmonic order 1 to 13 obtained with the TD-DFT calculation for the pentacene crystal (Fig. 3)
Yield_TB.txt	The first row contains the laser polarization, and the following rows contain the harmonic yield of harmonic order 1 to 13 obtained with the 2D-tight-binding model calculation (Fig. 3)
Yield_TB_flipped_t2_0_01.txt	The first row contains the laser polarization, and the following rows contain the harmonic yield of harmonic order 1 to 13 obtained with the 2D-tight-binding model calculation, that has been rotated, for a coupling $t_2 = 0.01$ (Extended Data Figure 3)
Yield_TB_flipped_t2_0_0001.txt	The first row contains the laser polarization, and the following rows contain the harmonic yield of harmonic order 1 to 13 obtained with the 2D-tight-binding model calculation, that has been rotated, for a coupling $t_2 = 0.0001$ (Extended Data Figure 3)

### 4.3 Tight\_Binding\_Flipping\_Analysis

Includes the data supporting the plots showing the  $t_2$  dependent polarization dependence of the modified tight-binding model, i.e. Fig. 4 and Extended Data Figure 3.

phi_of_maximum_yield.txt	Laser polarization $\varphi$ for which the harmonic yield is maximized as a function of $t_2$ for harmonic 1 - 13
t2_values.txt	Values of the tight-binding parameter $t_2$

### 4.4 Single\_HHG\_Spectra

Includes the data supporting the experimental HHG spectra shown in Fig.1 and the comparison of the individual HHG spectra shown in Extended Data Figure 2.

Spectrum_Experiment.txt	The first row contains the energy axis in units of $\omega/\omega_0$ , and the second row contains the experimentally measured harmonic spectra. Note, that the spectrum around the third harmonic was measured with a different spectrometer and has been shifted by a factor of 0.3. The electric field is polarized at an angle $\varphi = XXX^\circ$ , corresponding a polarization parallel to the direction of a nearest neighbor.
Spectrum_MR_CIS.txt	The first row contains the energy axis in units of $\omega/\omega_0$ , and the second row contains the harmonic yield obtained in the MR-CIS simulation for the individual molecules. The electric field is polarized at an angle $\varphi = 130^\circ$ , corresponding to a polarization parallel to the long molecular axis.
Spectrum_TD_DFT.txt	The first row contains the energy axis in units of $\omega/\omega_0$ , and the second row contains the harmonic yield obtained in the TD-DFT simulation for the crystal. The electric field is polarized at an angle $\varphi = 129.375^\circ$ , corresponding a polarization parallel to the direction of a nearest neighbor.

## References

- [1] Falk-Erik Wiechmann, Samuel Schöpa, Lina Bielke, Svenja Rindelhardt, Serguei Patchkovskii, Felipe Morales, Maria Richter, Dieter Bauer, Franziska Fennel, *High-Order Harmonic Generation in an Organic Molecular Crystal*, Nature Communications (submitted)