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Supplemental data for Investigation of Interferences in Carbon Dioxide through Multidimensional Molecular-Frame High-Harmonic Spectroscopy

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This documentation describes the time-dependent density-functional theory (TDDFT) data, and Python scripts, we use to make the theory figures published in "Investigation of Interferences in Carbon Dioxide through Multidimensional Molecular-Frame High-Harmonic Spectroscopy" [Tuthill 2022]. All the supplemental data were obtained with the Octopus package [Andrade 2012, Andrade 2015, Tancogne-Dejean 2020], version 8.4.

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2. Octopus-package license

3. Supplemental data types

There are two classes of file names: `acceleration_theta_deg=*` and `dipole_HOMO_theta_deg=*`, both with the same file format (note, however, that the former contains a five-line header, while the latter does not.) Each one of these files contains five columns: the first column indicates the iteration number (the number of time steps that have been performed up to that point in the simulation), the second column indicates the time (in atomic units), and columns 3-to-5 indicate the dipole signal in the x, y, and z directions, respectively. For more information about how these dipole signals are generated, see [Hamer 2021].

4. Supplemental data content

The supplemental data are sorted into 2 archive (zip) files:

- *Data:* contains the results of the harmonic-generation simulations with varying the molecular-alignment angle, used to create Figures 4 and 6 in the main text. The alignment angle is specified in each file name.
- *Scripts:* contains the scripts used to generate the figures seen in the paper and discussed in Section 4.

The "*Data*" archive contains three sub-directories with identical structures, labeled by their corresponding wavelengths: for each of the three wavelength/intensity combinations studied (800 nm, 3.5×10^{14} W/cm^2; 1500 nm, 6×10^{13} W/cm^2; 2000 nm, 6×10^{13} W/cm^2), there are 13 `acceleration_theta_deg=*` files, for angles 0, 5, 10… 60 degrees, containing the total dipole acceleration from the corresponding HHG simulation. Also, there are 13 `dipole_HOMO_theta_deg=*` files, for the same angles, containing the combined orbital-resolved dipole signals from the two degenerate highest-occupied molecular orbitals.

The "*Scripts*" archive contains two `.py` files. We have validated all the scripts on the Spyder IDE with Python 3.7.9.

- `TCI_analysis.py`: This script is the main analysis tool for high-harmonic spectroscopy computations within Octopus. When given a directory containing the output of several angle-dependent Octopus calculations, it computes the (1) spectral intensity, (2) spectral phase, and (3) target-specific group delay, and then it plots them versus the angle between the laser and molecular axis. The selection of the plot is controlled with three global variables: `wl` (the wavelength), `cwd` (the file path where the relevant directory is located), and `orbital_resolved` (a Boolean, indicating whether to plot the `acceleration` or `total_HOMO` signal).
- `utils.py`: This script contains methods that are used throughout all of the scripts. Make sure that your integrated development environment (IDE) can locate `utils.py`.

5. Acknowledgments

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6. References

[Andrade 2012] X. Andrade, J. Alberdi-Rodriguez, D.A. Strubbe, M.J.T. Oliveira, F. Nogueira, A. Castro, J. Muguerza, A. Arruabarrena, S.G. Louie, A. Aspuru-Guzik, A. Rubio, and M.A.L. Marques, "Time-dependent density-functional theory in massively parallel computer architectures: the octopus project," J. Phys.: Condens. Matter 24, 233202 (2012).

[Andrade 2015] X. Andrade, D. Strubbe, U. De Giovannini, A.H. Larsen, M.J.T. Oliveira, J. Alberdi-Rodriguez, A. Varas, I. Theophilou, N. Helbig, M.J. Verstraete, L. Stella, F. Nogueira, A. Aspuru-Guzik, A. Castro, M.A.L. Marques, and A. Rubio, "Real-space grids

and the Octopus code as tools for the development of new simulation approaches for electronic systems," Phys. Chem. Chem. Phys. 17, 31371 (2015).

[Hamer 2021] K.A. Hamer, D.R. Tuthill, T.D. Scarborough, L.F. DiMauro, K. Lopata, K.J. Schafer, M.B. Gaarde, and F. Mauger, "Orbital-resolved calculations of two-center interferences in linear triatomic molecules," Phys. Rev. A 104, 033114 (2021).

[Tancogne-Dejean 2020] N. Tancogne-Dejean, M.J.T. Oliveira, X. Andrade, H. Appel, C.H. Borca, G Le Breton, F. Buchholz, A. Castro, S. Corni, A.A. Correa, U. De Giovannini, A. Delgado, F.G. Eich, J. Flick, G. Gil, A. Gomez, N. Helbig, H. Hübener, R. Jestädt, J. Jornet-Somoza, A.H. Larsen, I.V. Lebedeva, M. Lüders, M.A.L. Marques, S.T. Ohlmann, S. Pipolo, M. Rampp, C.A. Rozzi, D.A. Strubbe, S.A. Sato, C. Schäfer, I. Theophilou, A. Welden, and A. Rubio, "Octopus, a computational framework for exploring light-driven phenomena and quantum dynamics in extended and finite systems," J. Chem. Phys. 152, 124119 (2020).

[Tuthill 2022] D.R. Tuthill, T.D. Scarborough, T.T. Gorman, K.A. Hamer, R.R. Jones, M.B. Gaarde, K. Lopata, F. Mauger, K.J. Schafer, and L.F. DiMauro, "Investigation of Interferences in Carbon Dioxide through Multidimensional Molecular-Frame High-Harmonic Spectroscopy," J. Phys. Chem. A 126, 8588 (2022).