Supplemental Document

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Detecting multiple chiral centers in chiral molecules with high harmonic generation: supplement

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Supplemental Material: Detecting multiple chirality centers in chiral molecules with high harmonic generation

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1. METHODS

DFT calculations were performed with OCTOPUS code [1–3]. The KS equations were discretized in a spherical box with radius 34 Bohr with a Cartesian grid, where molecular centers of mass were centered at the origin. Calculations were performed within the local density approximation (LDA) with an added self-interaction correction (SIC) [4]. The frozen core approximation was used for inner core orbitals, which were treated with norm-conserving pseudopotentials [5]. The KS equations were solved to self-consistency with a tolerance $<10^{-7}$ Hartree, and the grid spacing was converged to 0.4 Bohr. All molecular structures were relaxed $<10^{-4}$ Hartree/Bohr in forces within the LDA.

For TDSE calculations as described in the main text, we utilized a time step Δt=0.11 a.u. with an imaginary absorbing potential of width 8 Bohr at the boundary. The grid size, absorbing potential, and time step were tested for convergence.

2. Three-Center Multi-Chiral System

We present here sketches of the molecular geometries of the utilized three-center multi-chiral system exported in the main text, C_5H_9BrClF. Fig. S1 presents the stereo-chemical relationships between the different isomers.

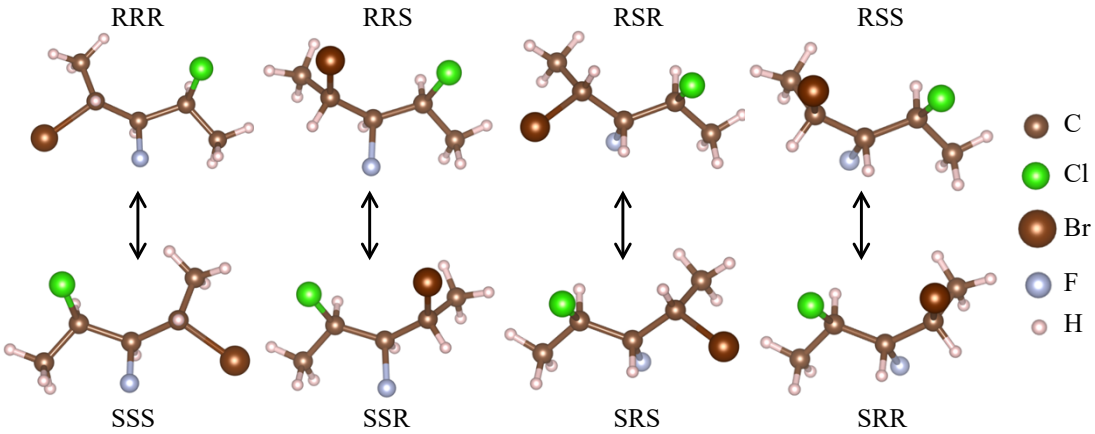

FIG. S1. Schematic illustration of the three-center chiral system showing all eight stereoisomers of chiral molecules for C_5H_9BrClF. The arrows indicate enantiomeric pairs of molecules that are mirror images of one another, while diastereomers are not labeled. Labeling of the chiral center is around the three carbons in the chain, respectively, e.g. 'RSR' labels a chiral molecule with 'R' configuration around the 1st carbon, and 'S' around the 2nd, and 'R' around the 3rd.

REFERENCES

1. A. Castro, H. Appel, M. Oliveira, C. A. Rozzi, X. Andrade, F. Lorenzen, M. A. L. Marques, E. K. U. Gross, and A. Rubio, "octopus: a tool for the application of time-dependent density functional theory," Phys. status solidi **243**, 2465–2488 (2006).
2. X. Andrade, D. Strubbe, U. De Giovannini, H. Larsen, M. J. T. Oliveira, J. Alberdi-rodriguez, A. Varas, I. Theophilou, N. Helbig, M. J. Verstraete, L. Stella, F. Nogueira, 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**, 31371–31396 (2015).
3. 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).
4. J. P. Perdew and A. Zunger, "Self-interaction correction to density-functional approximations for many-electron systems," Phys. Rev. B **23**, 5048–5079 (1981).
5. C. Hartwigsen, S. Goedecker, and J. Hutter, "Relativistic separable dual-space Gaussian pseudopotentials from H to Rn," Phys. Rev. B **58**, 3641–3662 (1998).