Bond Ellipticity Alternation: An Accurate Descriptor of Non-linear Optical Properties of π-Conjugated Chromophores

Thiago O. Lopes,[†] Daniel F. Scalabrini Machado,[†] Chad Risko,[§] Jean-Luc Brédas[,‡] and Heibbe C. B. de Oliveira[*,†]*

[†] Laboratório de Estrutura Eletrônica e Dinâmica Molecular (LEEDMOL), Institute of Chemistry, University of Brasília, Campus Darcy Ribeiro, Brasília, Brazil.

[§] Department of Chemistry and Center for the Applied Energy Research, University of Kentucky, Lexington, Kentucky 40506-0055, United States of America.

[‡] School of Chemistry and Biochemistry and Center for Organic Photonics and Electronics, Georgia Institute of Technology, Atlanta, Georgia 30332-0400, United States of America.

Corresponding Authors

*Email: jean-luc.bredas@chemistry.gatech.edu

*Email: heibbe@unb.br

Table of Contents:

A. Additional results for the 5-carbon streptocyanine
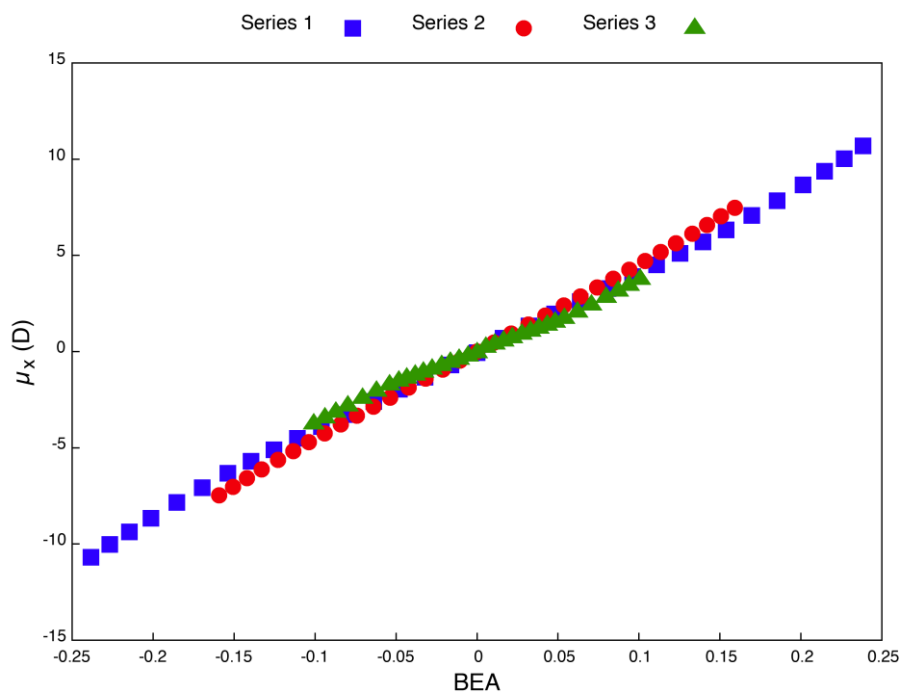
A1. Dipole Moment

Figure S1. Correlation of μ_x with BEA for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
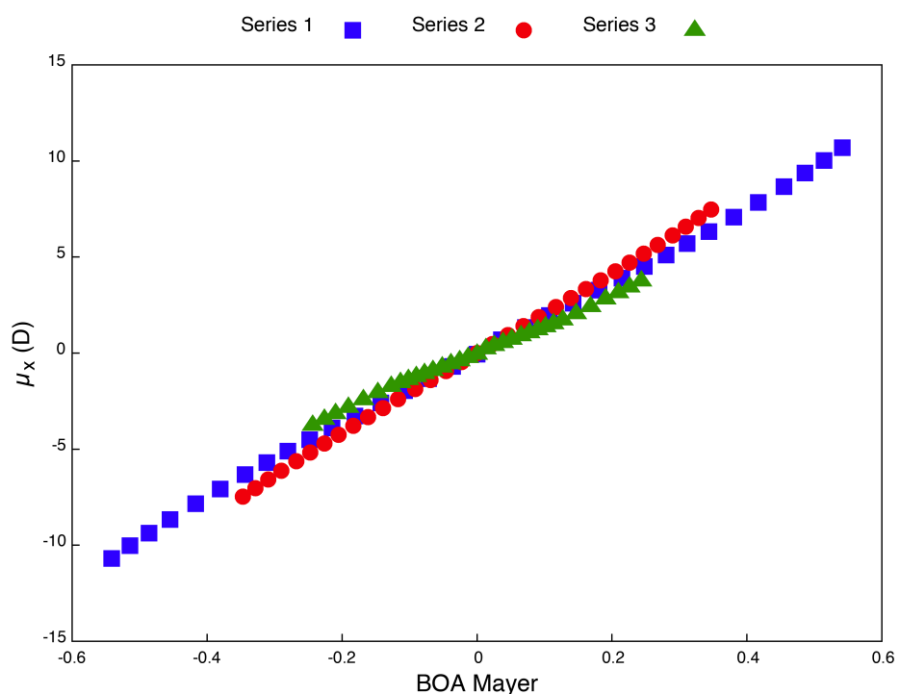
Figure S2. Correlation of μ_x with BOA Mayer for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
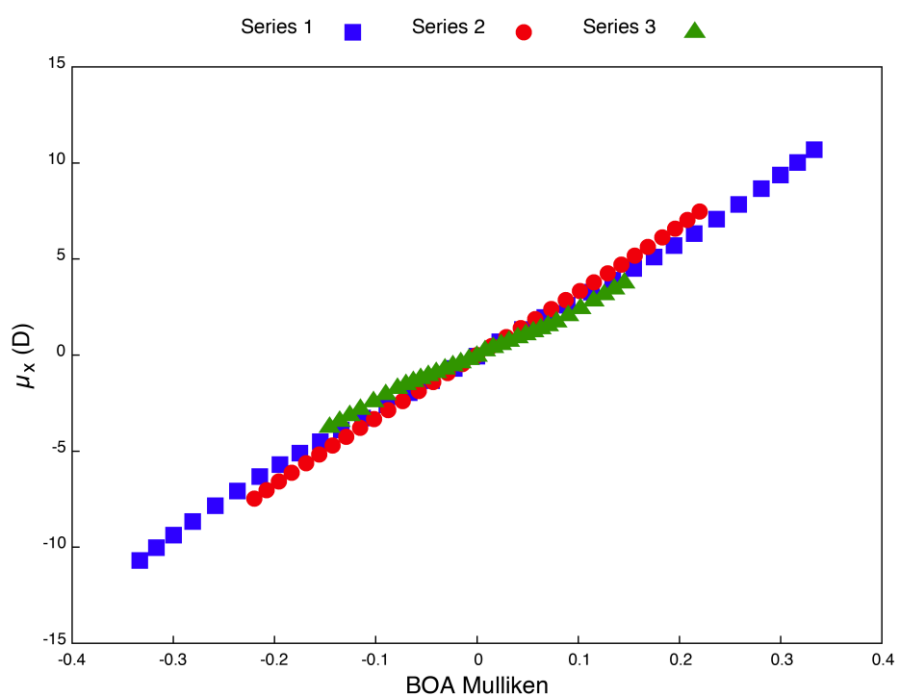

Figure S3. Correlation of μ_x with BOA Mulliken for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
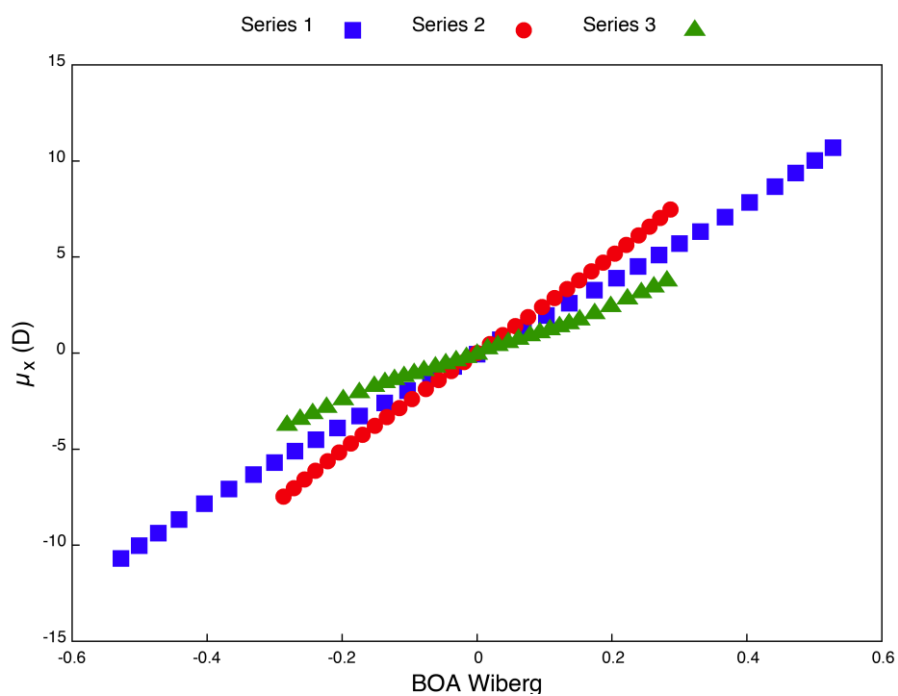
Figure S4. Correlation of μ_x with BOA Wiberg for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
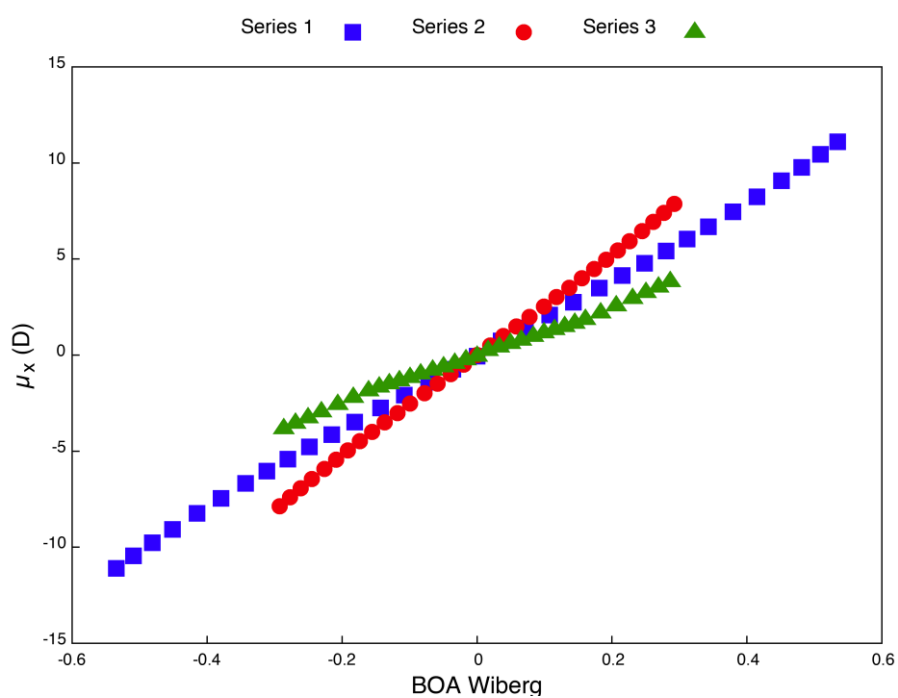

Figure S5. Correlation of μ_x with BOA Wiberg for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31++G(d, p) level of theory.
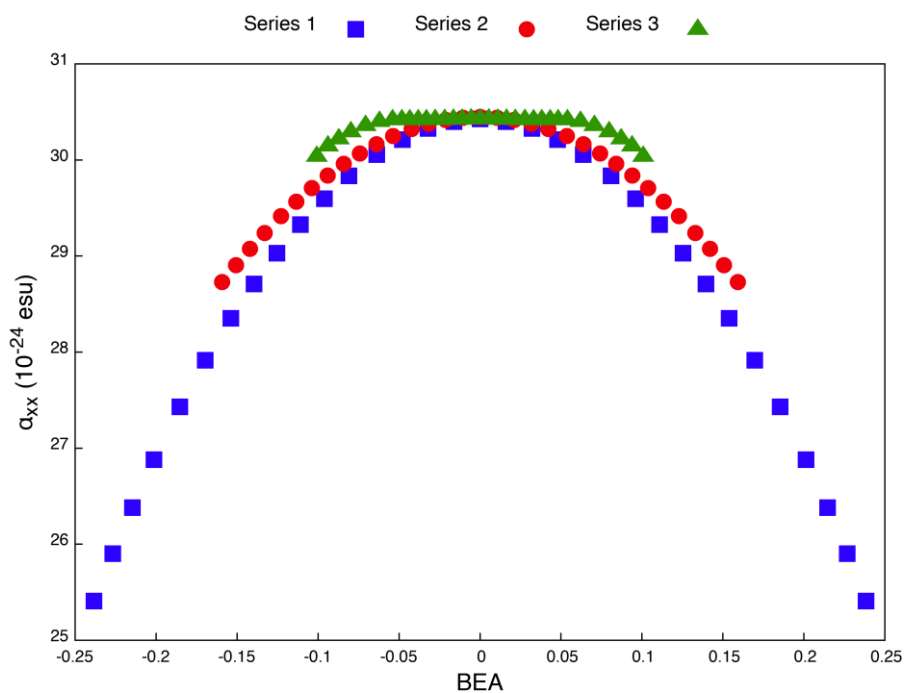
A2. First-Order Polarizability

Figure S6. Correlation of α_{xx} with BEA for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
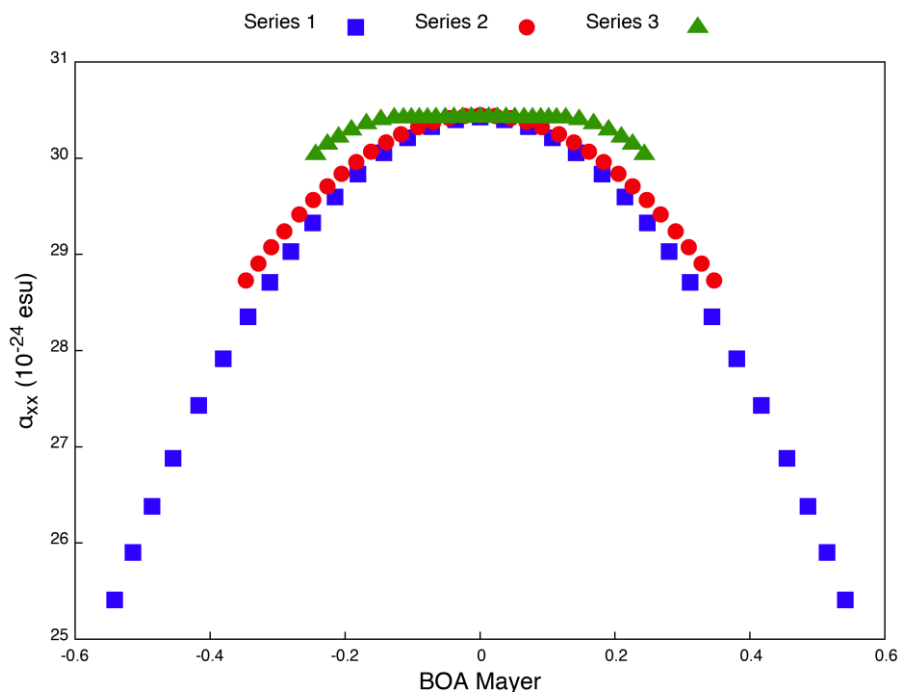

Figure S7. Correlation of α_{xx} with BOA Mayer for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
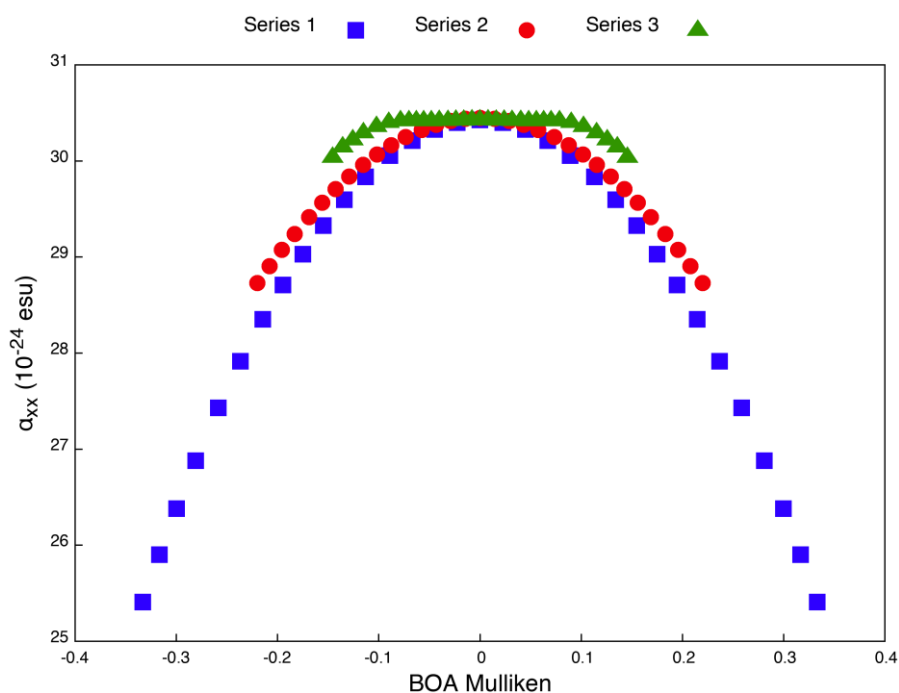
Figure S8. Correlation of α_{xx} with BOA Mulliken for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
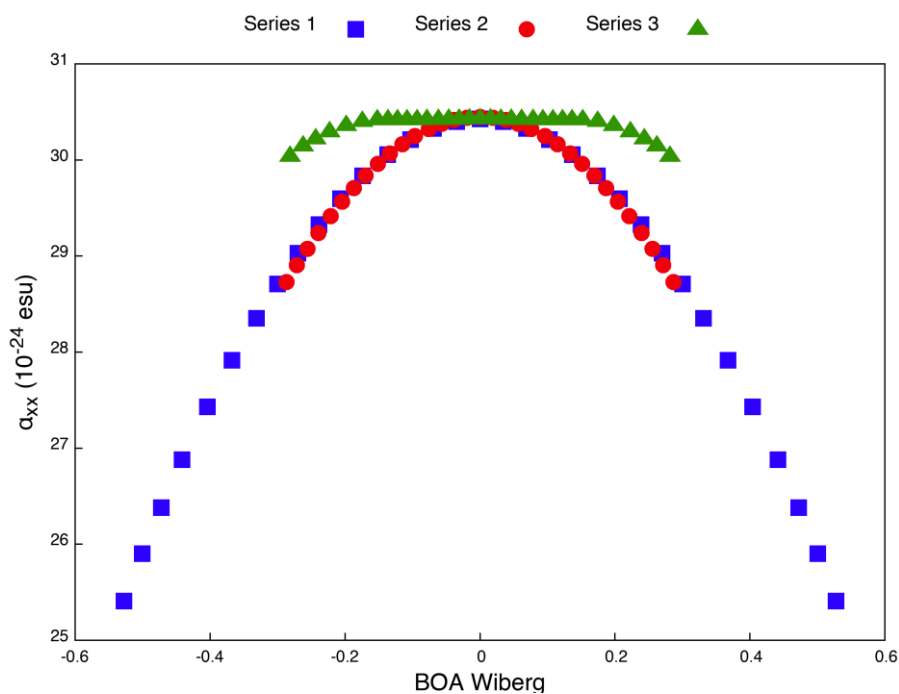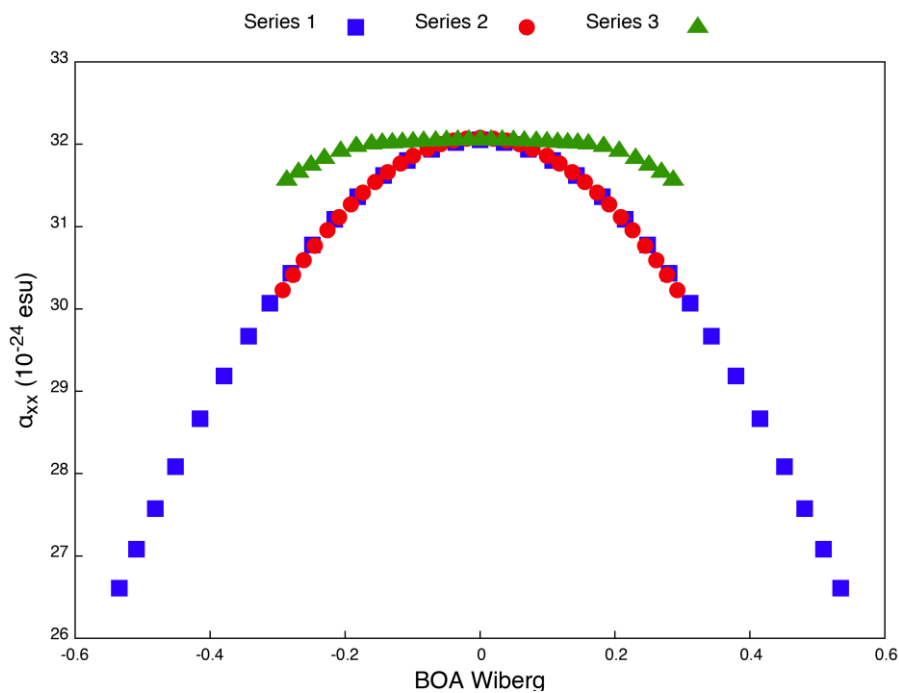

Figure S9. Correlation of α_{xx} with BOA Wiberg for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.

Figure S10. Correlation of α_{xx} with BOA Wiberg for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31++G(d, p) level of theory.
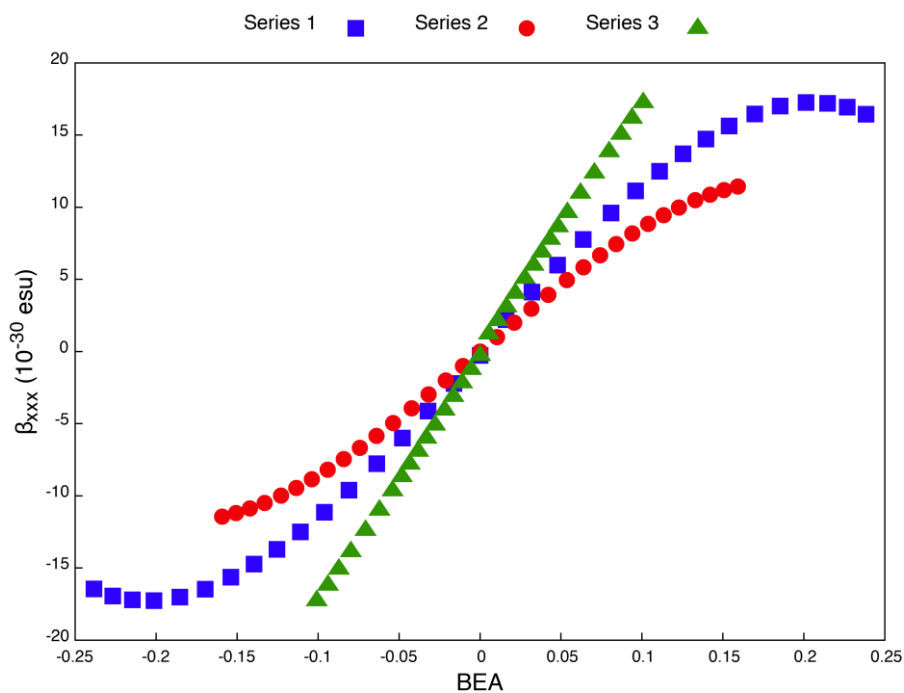
A3. Second-Order Polarizability

Figure S11. Correlation of β_{xxx} with BEA for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
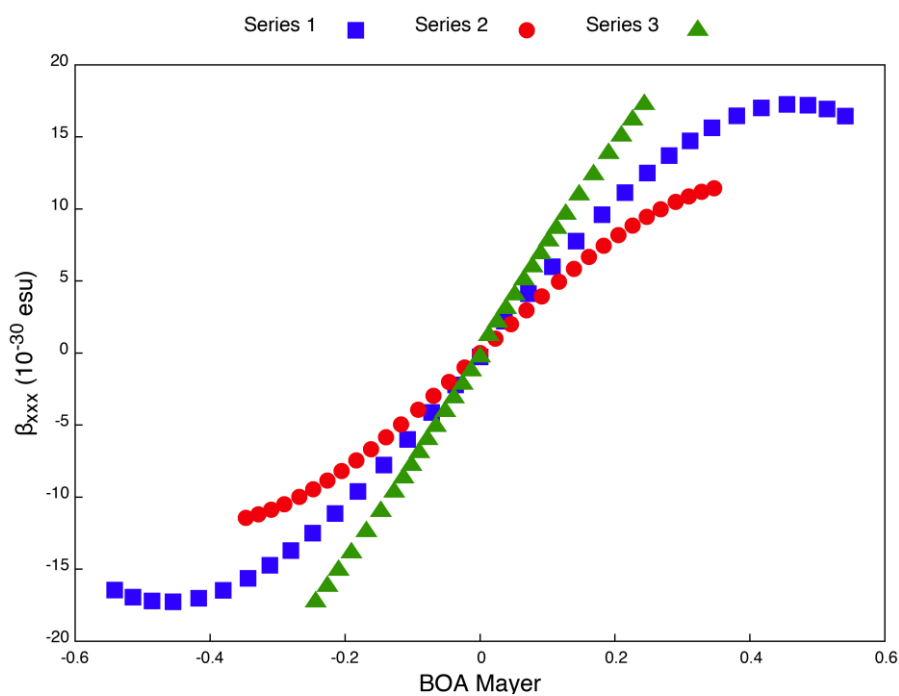
Figure S12. Correlation of β_{xxx} with BOA Mayer for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
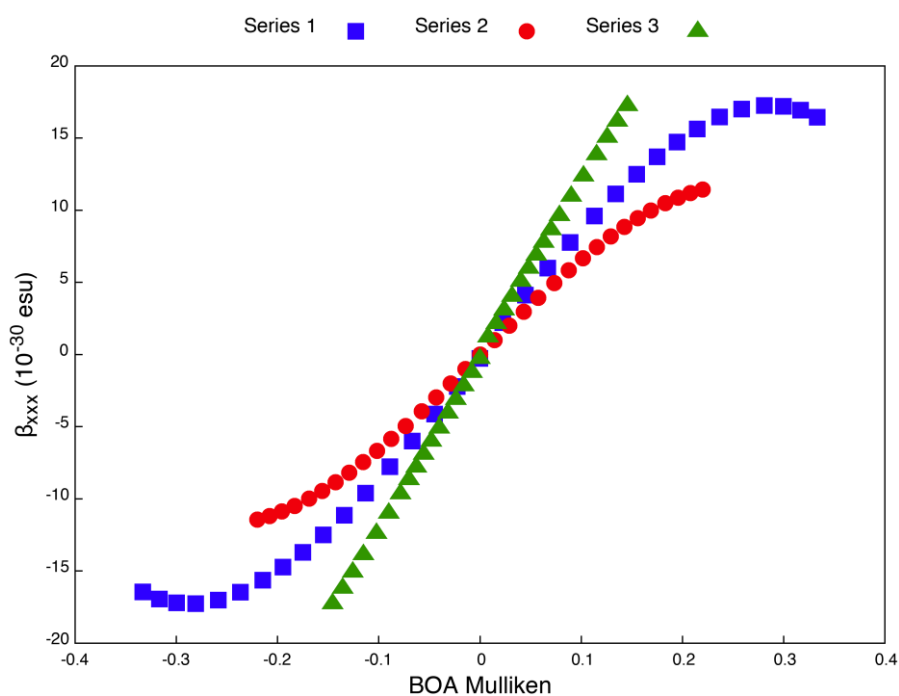

Figure S13. Correlation of β_{xxx} with BOA Mulliken for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
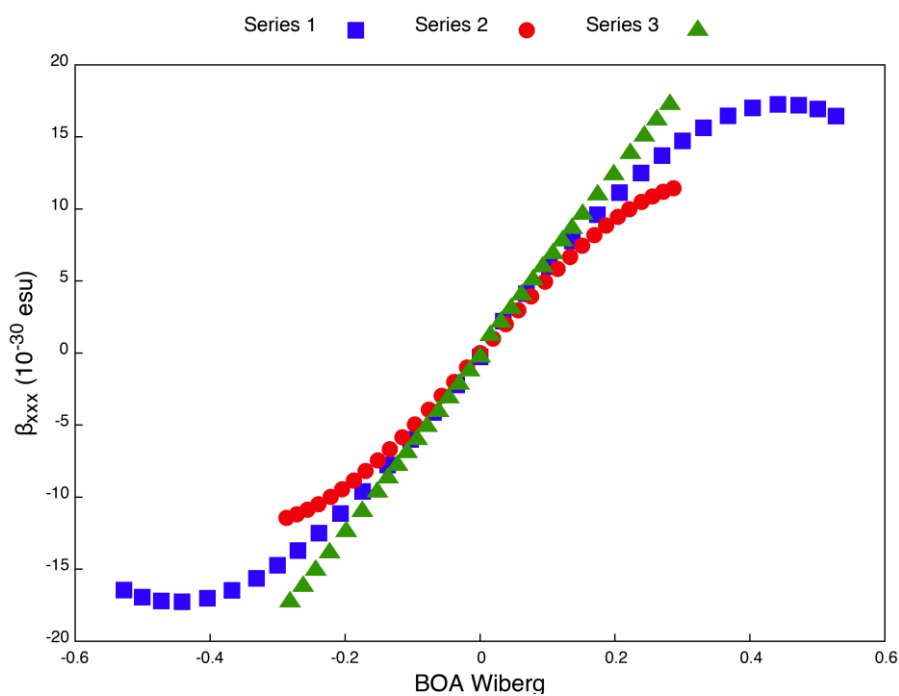
Figure S14. Correlation of β_{xxx} with BOA Wiberg for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
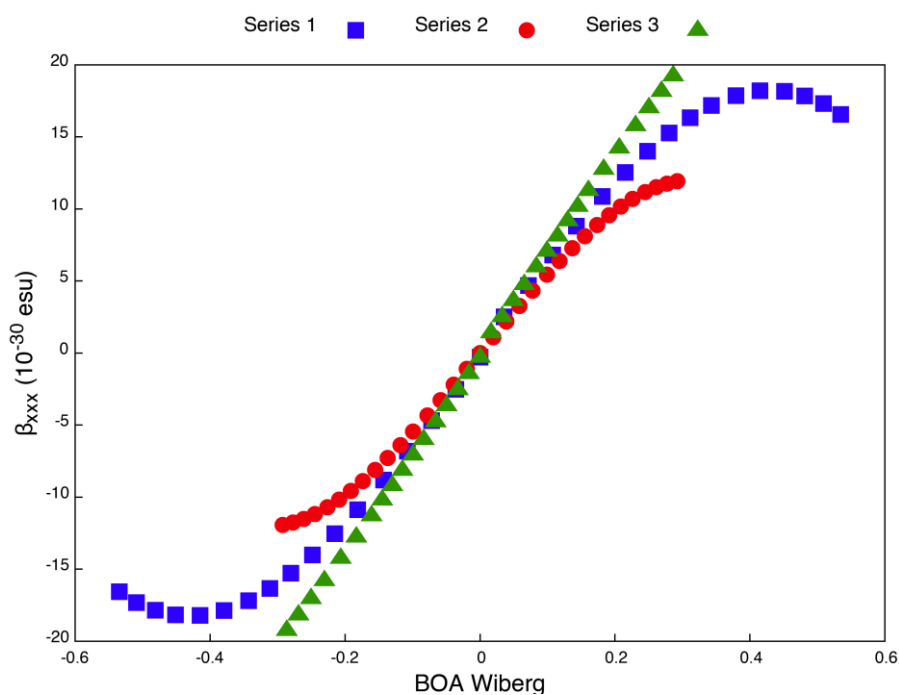

Figure S15. Correlation of β_{xxx} with BOA Wiberg for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31++G(d, p) level of theory.
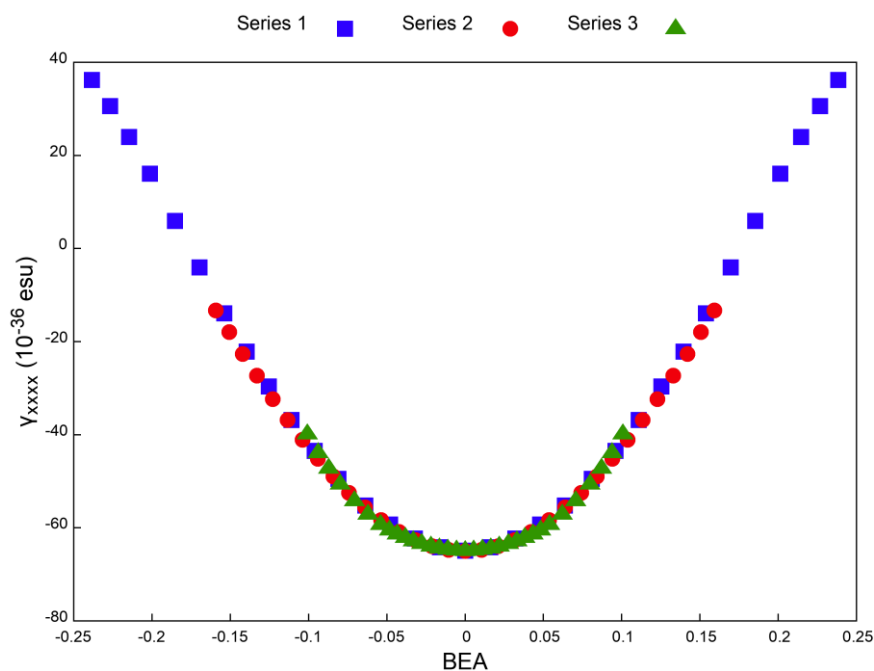
A4. Third-Order Polarizability

Figure S16. Correlation of γ_{xxxx} with BEA for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
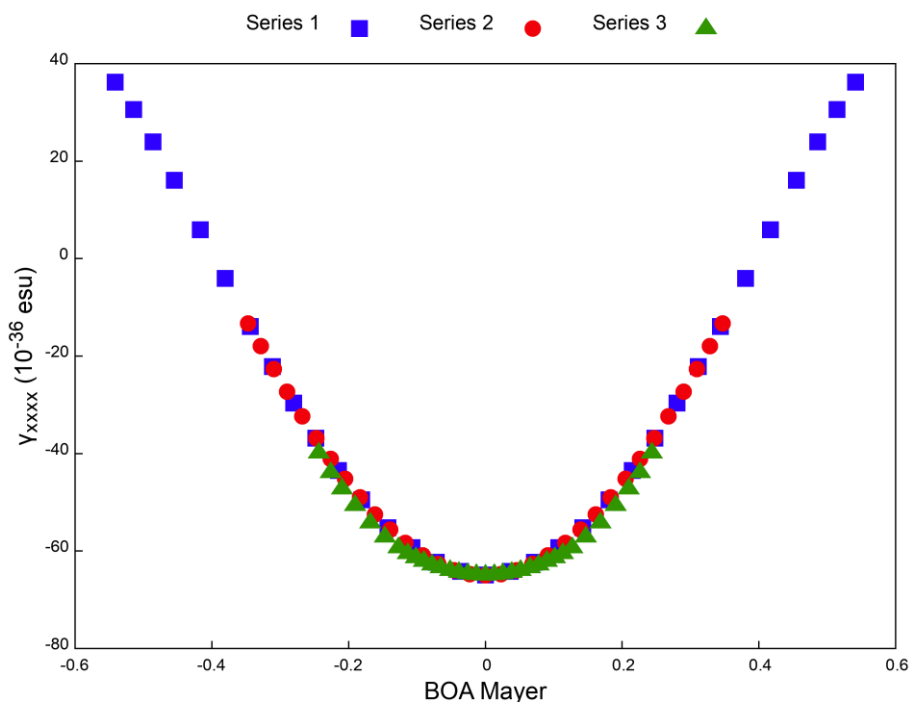

Figure S17. Correlation of γ_{xxxx} with BOA Mayer for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
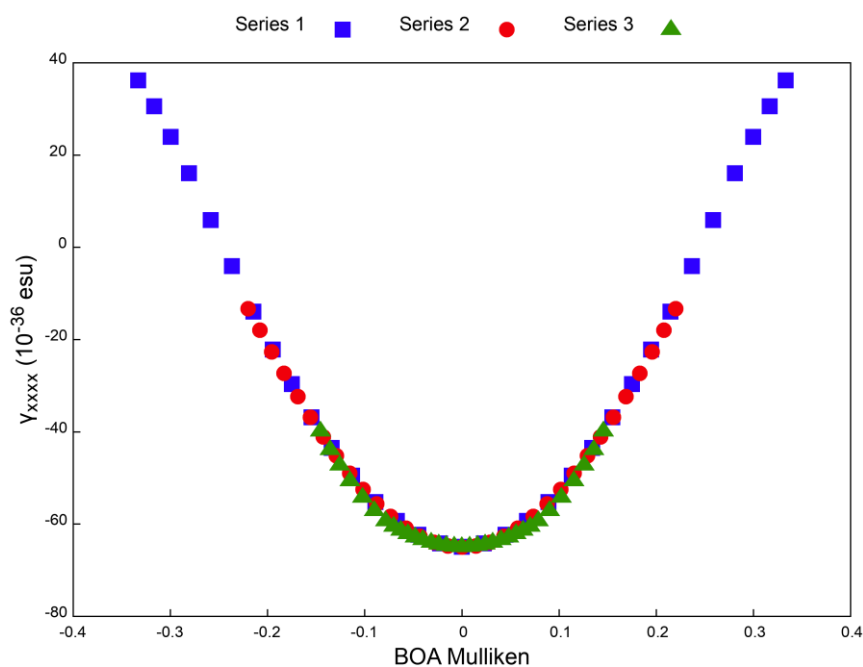
Figure S18. Correlation of γ_{xxxx} with BOA Mulliken for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
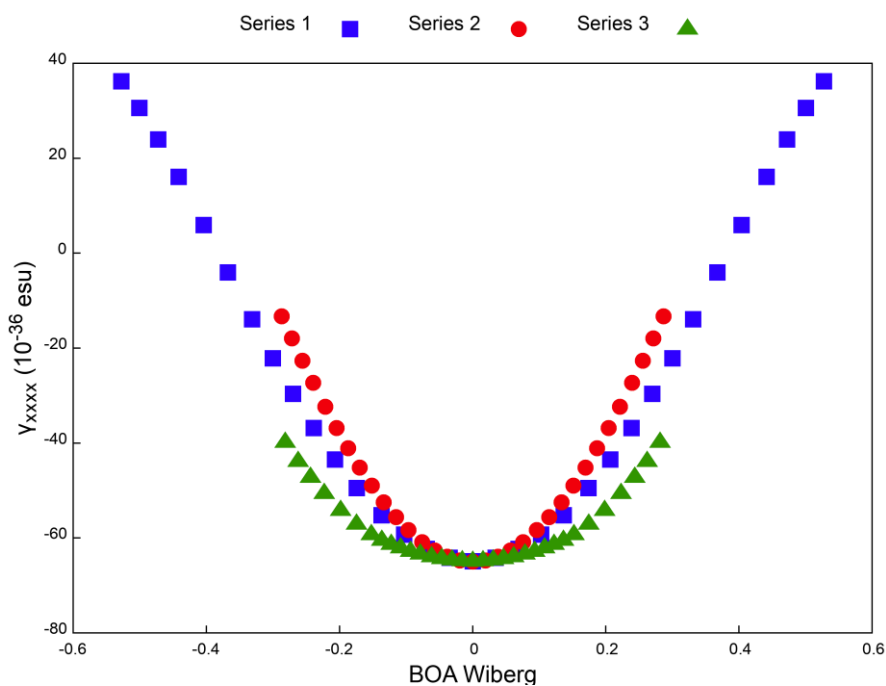

Figure S19. Correlation of γ_{xxxx} with BOA Wiberg for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31G(d, p) level of theory.
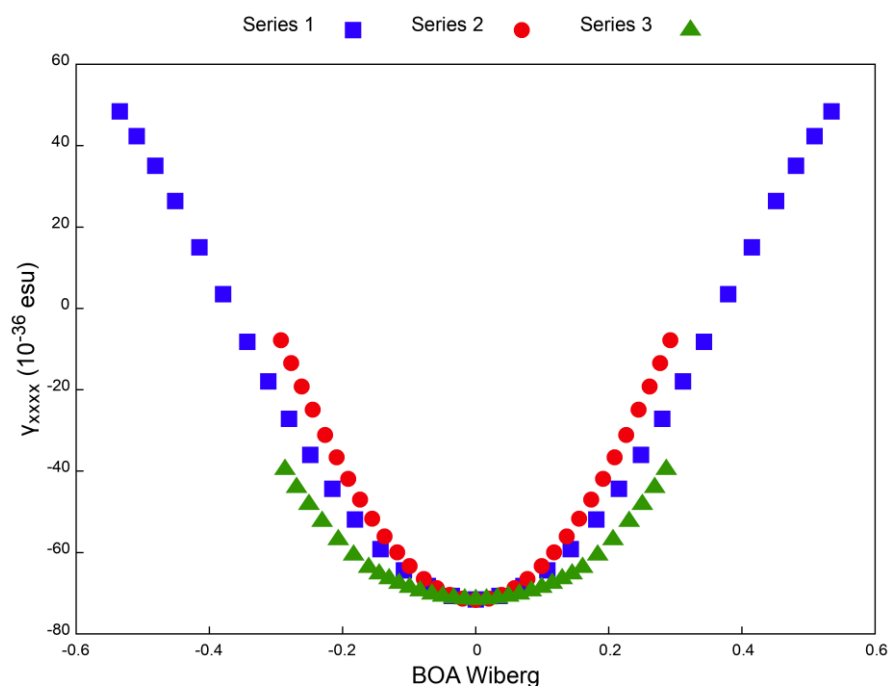
Figure S20. Correlation of γ_{xxxx} with BOA Wiberg for the 5C streptocyanine in Series 1, 2, 3, as obtained at the ωB97XD/6-31++G(d, p) level of theory.