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Research Article

The Solute Polarization and Structural Effects on the Nonlinear Optical Response of Based Chromone Molecules

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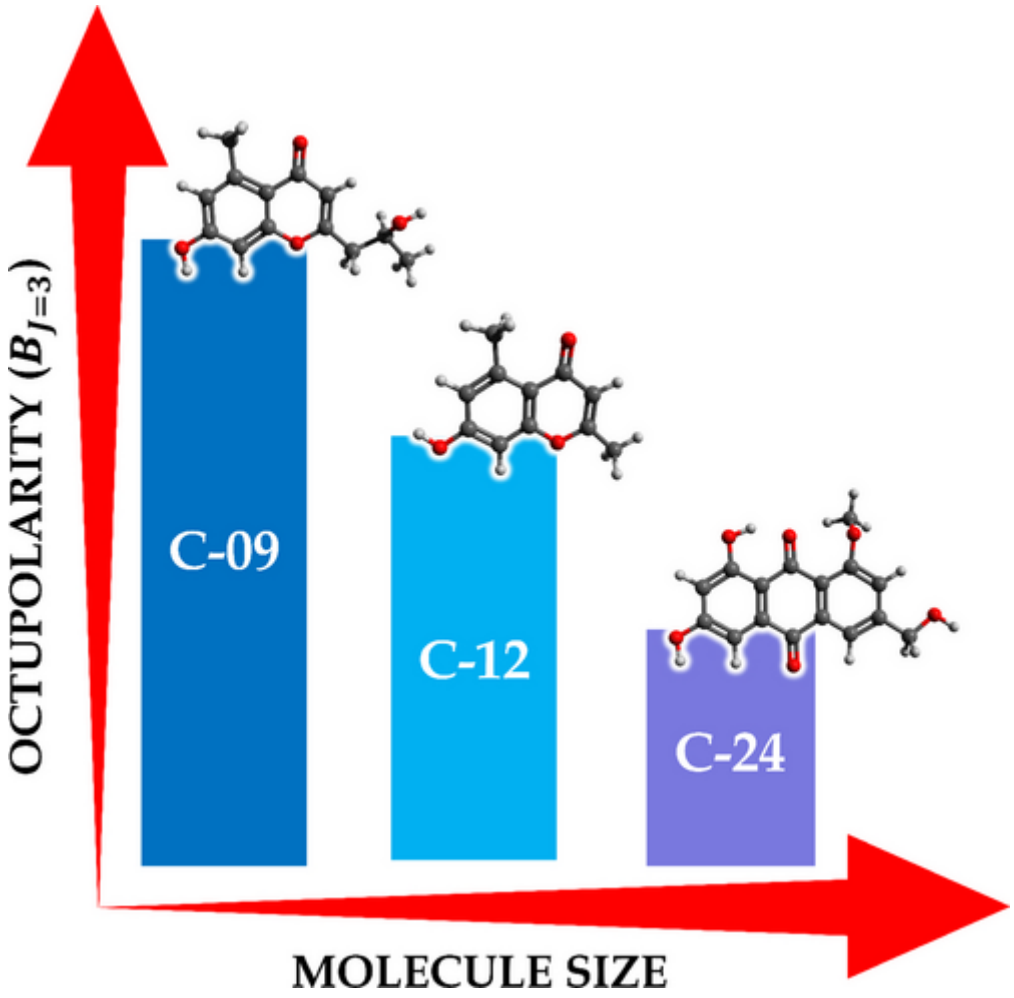
Graphical Abstract

Sum of dipolar ($\Phi_{J=1}$) and octupolar ($\Phi_{J=3}$) contributions determine the nonlinear optical (NLO) behavior in a material. Octupolar chromophores are rare but essential in NLO and thus, it is crucial to learn how to balance these characteristics. The increase of the molecule size in a preferential direction and the polarization of the solute due to the solvent decreases the octupolar contributions in favor of the dipolar one.



Abstract

The solute polarization due to solvent is a an electrostatic quantum effect that impacts diverse molecular properties, including the nonlinear optical response of a material. An iterative procedure that allows updating the solute charge distribution in the presence of the solvent is combined with a sequential Monte Carlo/Quantum Mechanics methodology and Density Functional Theory methods to evaluate the nonlinear optical (NLO) response using the hyper Rayleigh scattering (HRS) of a series of chromones recently identified in *Chamaecrista diphylla*, an herbaceous plant abundant throughout the Americas and used in folk

medicine. From this study, it is determined that from gas to solvent environment, the systems acquire low refractive index (n) and an improvement of the first hyperpolarizability (β_{HRS}), signaling potential NLO uses. It is shown that the octupolar contributions ($\beta^{J=3}$) superate the dipolar ones ($\beta^{J=1}$) and dominate the second-order optical response in both gas and liquid phases, which indicate nontrivial optical materials. Moreover, the solvent environment and structural changes in the periphery can tune significantly the dipolar/octupolar balance, showing a key to control the decoupling between these contributions.

Conflict of interest

There is not conflict of interest.

Open Research

Data Availability Statement

All the data for this researche is already available in the submitted manuscript.

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