



SDI Review Form 1.6

Journal Name:	<a href="#">Asian Journal of Chemical Sciences</a>
Manuscript Number:	Ms_AJOCS_41290
Title of the Manuscript:	The static (hyper)polarizabilities of push-pull polyenes : A theoretic and computational study
Type of the Article	

**General guideline for Peer Review process:**

This journal's peer review policy states that **NO** manuscript should be rejected only on the basis of '**lack of Novelty**', provided the manuscript is scientifically robust and technically sound.

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**PART 1: Review Comments**

	Reviewer's comment	Author's comment (if agreed with reviewer, correct the manuscript and highlight that part in the manuscript. It is mandatory that authors should write his/her feedback here)
<b>Compulsory</b> REVISION comments	<p>In the manuscript the authors studied the (hyper)polarizabilities of push-pull polyenes. For a series of NH<sub>2</sub>-(CH=CH)<sub>N</sub>-NO<sub>2</sub> molecules with growing N (from 2 to 6), their (hyper)polarizabilities were calculated at 6 levels of theory. Changes in the calculated (hyper)polarizabilities as a function of N and the level of theory were tracked and, on this basis, some conclusions on the accuracy of the used levels of theory were drawn.</p> <p>1. The manuscript is not well written. Its length is insufficient for the inclusion of all relevant details and making a meaningful discussion. Several findings were described superficially, and in consequence, unclearly.</p> <p>2. The computational protocol used in the study is not justified. The application of HF/6-31G for optimization of small molecules (the number of atoms in the largest molecule does not exceed 30 atoms) is far below today's standard. The range of computational methods used in the study is also rather narrow (only 6 methods were included). The authors should include more density functionals (see for example Journal of Computational Chemistry 2013, 34, 819–826). Another important issue in computational studies of (hyper)polarizabilities is a careful choice of basis set. Both 6-31G(d) and cc-pVDZ were not designed to predict hyper(polarizabilities) accurately.</p> <p>3. The authors assessed the performance of computational methods in predicting the (hyper)polarizabilities of NH<sub>2</sub>-(CH=CH)<sub>N</sub>-NO<sub>2</sub>. However, it is not clear what reference results were used for this assessment. If CCSD(T) results were used as reference, then there are only part of such calculations in the manuscript (for N = 2,3,4). In the case of CCSD(T), an issue regarding the heavy incompleteness of basis set occurs. Furthermore, the authors did not quote any experimental results (if there are none, they should stress it).</p>	<p>We agree with the reviewer, and correct the manuscript as required.</p> <p>1. We checked thoroughly the manuscript and improved the language of the manuscript. We added the data used in the plots. The discussion and conclusion parts were carefully revised.</p> <p>2. We added comments on calculations reported by other researchers, which meanwhile justifies or rationalizes the computational protocol used in the manuscript. Conventional DFT methods were known to be unreliable, so we just choose one typical functional B3LYP. The 6-31G(d) and cc-pVDZ are insufficient to predict response properties accurately, and we focus the performances of the electronic structure methods while response properties of push-pull polyenes are studied in this paper.</p> <p>3. CCSD results are used as reference. We revised the manuscript to make this point more clear. It is difficult to compare theoretic results with experimental data (partly due to the absence of the experimental data). No experimental results were quoted in similar publications (eg. Ref. 8-14).</p>
<b>Minor</b> REVISION comments	<p>1. Symbols in Eqs. 1-3 should be explained.</p> <p>2. Eq. 2 is incorrect: E(h) → E(-h).</p> <p>3. References to the finite field method should be added.</p> <p>4. A table with the reference and extrapolated values of (hyper)polarizabilities would be very much appreciated.</p> <p>5. In the abstract the authors did not mention the CCSD(T) method but this method was used in the study.</p> <p>6. A brief review of previous theoretical studies of push-pull polyenes should be included in the introduction.</p>	<p>1. Symbols used here were explained in the revised manuscript.</p> <p>2. Eq. 2 was corrected.</p> <p>3. References to finite field method were added (Ref. 21-25).</p> <p>4. We added three tables to give the relevant data.</p> <p>5. Since the CCSD(T) calculations were carried out only for a part of systems studied in the manuscript and CCSD results were used as reference, we didn't mention the CCSD(T) method in the abstract.</p> <p>6. We added comments on previous theoretical studies in the introduction.</p>
<b>Optional/General</b> comments	<p>1. I advise the authors to change the title slightly: 'A theoretic and computational study' → 'A theoretical study'.</p> <p>2. There are several grammar mistakes in the manuscript.</p>	<p>1. We accept the advice. The title has been revised.</p> <p>2. We have checked the manuscript and corrected several grammar mistakes.</p>