Development of efficient geometry optimization method at ground and excited states towards large systems

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（大規模系に向けた基底状態および励起状態の効率的構造最適化法の開発）

Title

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論　文　内　容　の　要　旨

Thesis Summary

This thesis introduces a novel approach in computational chemistry for optimizing the geometry of ground and excited states in large molecules, such as proteins and DNA. Focusing on overcoming the prohibitive computational costs associated with conventional methods, this research improves the Elongation (ELG) method by combination of electrostatic and mechanical embeddings and significantly modifies the Local Excitation Approximation (LEA) method.

The ELG, a method for simulation of polymers, calculates molecular regions sequentially, facing accuracy issues due to ignoring long-range interactions. This research introduces electrostatic and mechanical embeddings to address this. Electrostatic embedding uses atomic charges from Charge Sensitivity Analysis (CSA), parametrical method for efficient calculation of charges. Mechanical embedding includes cost-effective methods for subsequent regions, improving the initial geometries for later steps. These enhancements are tested on polyethylene and nylon chains.

The LEA method, effective for calculating localized electronic excitations in optically active center of molecules, has been significantly modified. The modifications include expanding the number of Regional Localized Molecular Orbitals (RLMOs) and refining localized canonical molecular orbitals (CMOs) using an “updating” technique. Tested on various systems, including a green fluorescent protein, the modified LEA method showed higher accuracy than original method keeping computational efficiency.

Obtained in this research new models for CSA in combination with ELG significantly enhances electronic energy accuracy in evaluation calculations with proteins and DNA. The combined electrostatic and mechanical embeddings generally improve results but vary between polyethylene and nylon systems.

Besides, the modified LEA method delivers highly accurate excitation energy calculations, demonstrating at least quadratic acceleration in calculation speed. Developed method also demonstrated high accuracy and efficiency in geometry optimization of excited state in polycarbonates.

This research has profound implications for computational chemistry. Developed methods open new possibilities in areas like drug discovery and material science. However, the varying outcomes observed across different systems highlights the necessity for further study. Future research will include this objective, with a focus on the combination of the developed methods for the study of large molecules.