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論文 / 著書情報
Article / Book Information

題目(和文)	振動分光および生体金属分子系の計算を目指したＦＭＯ法の開発
Title(English)	Development of Fragment Molecular Orbital Method toward the Application to Vibration Spectra and Inorganic Biochemistry
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出典(和文)	学位:博士（工学）, 学位授与機関:東京工業大学, 報告番号:甲第9811号, 授与年月日:2015年3月26日, 学位の種別:課程博士, 審査員:櫻井　実,藤井　正明,小長谷　明彦,田口　英樹,大谷　弘之,中村　振一郎
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学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

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（博士課程）
Doctoral Program

論 文 要 旨

THESIS SUMMARY

専攻： Department of	生体分子機能工学　専攻	申請学位（専攻分野）： Academic Degree Requested	博士 Doctor of　（　工学　）
学生氏名： Student's Name	中田　浩弥	指導教員（主）： Academic Advisor(main)	櫻井　実
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

Recently, the quantum chemistry is applied to large system due to the rapid increase of the computer resources. However, the methodology is limited toward applying to the biological systems, because the computational cost increases steeply with the system size. Thus, new efficient theory and computer program for solving the biological system are necessary. There are mainly two approaches for the calculation of large biological molecules, linear scaling approach and fragmentation approach.

The fragment molecular orbital method (FMO) is one of the fragmentation approaches. In FMO, the system is divided into the small subsets of fragments, and each fragment (monomer) is calculated independently in a presence of the electrostatic potential (ESP). The ESP is self-consistently obtained by iterative calculation of the SCF for each monomer. After the self-consistent ESP is obtained, the fragment pair (dimer) calculations are performed, and various physical properties can be estimated by the linear combination of monomers and dimers. Due to the fragmentation of the system, the FMO achieves nearly liner scaling calculation with the system size, and the calculations of the system possessing more than ten thousand atoms are possible.

However, the applications were mostly limited within the closed-shell system for the energy and optimization, and analytic second order derivative of the energy was not developed. Thus, reaction path analysis for metal enzyme protein was not possible. The second order derivative of the energy also can calculate the IR spectrum and Raman scattering, thus the energy second order derivative is very useful.

This study presents the new development of theory and methodology for the large biological systems, especially focusing on the metal enzyme reactions and vibration spectroscopy in biochemistry. For this purpose, analytic energy gradients for FMO-UHF and FMO-UDFT have been developed in this study, and they have been applied to predict the absorption spectrum of metal protein. Secondly, the analytic second order derivative of the energy has been developed for both restricted and unrestricted Hartree-Fock method, and applied to Raman spectra of biology.

Therefore, this thesis mainly consists of six chapters, and the former three chapters focus on the development of analytic energy gradient of open-shell systems. Therein, the accuracy of analytic energy gradient is evaluated by taking a comparison with numerical energy gradient, and the computational timing is demonstrated (Chapter 2 and 3). Secondary, the method is applied to predict the absorption spectra of metal protein (Chapter 4). The drastic improvement for accuracy of analytic energy gradient has been achieved, and reasonable agreement with experimental absorption spectra has been obtained. The latter three chapters are the development of the second order derivative of energy for FMO (FMO Hessian). First, the formulation of closed-shell systems is discussed, and the performance is evaluated by a comparison with full *ab initio* calculations (Chapter 5). Secondly, the method is extended to open-shell system, and the protein IR spectra are evaluated (Chapter 6). Finally, the FMO Hessian is applied to calculate Raman spectra for the protein consisting of more than hundreds atoms (Chapter 7).

Summarizing, FMO method is interfaced with open-shell method, and analytic energy gradient and the excited state calculation become possible. Additionally, the analytic second order derivative of the energy is formulated for both closed and open shell systems, which enables us to evaluate the reaction free energy and vibration spectra for large biological proteins. This development will open the new application territory of quantum chemistry, and we hope that the method will be widely used for the analysis of the reaction free energy and prediction of various spectroscopic properties.

備考 ： 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。
Note : Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1copy of 800 Words (English).

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