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論　文　内　容　要　旨

（NO. 1）

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学位論文の 題　　目	Theory of optical responses in multi-orbital systems with strong electron correlation (強相関多軌道系における光学応答の理論)		

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II. Quantum Cluster Calculation
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5. Electronic states and optical responses in Excitonic Insulating System
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6. Summary

Multi-orbital systems with strong electron correlation give rise to novel physics which can not be understood by single particle pictures. To fully understand the physics in those systems is of great significance but also a tough work. Experimentally, optical conductivity spectra are one of the most powerful methods to explore the electron structure in the correlated material. Two prototypical systems of the multiorbital correlated electron systems have attracted much attention, the dimer-type organic molecular system and the excitonic insulating system.

Dimer-type organic molecular solids exhibit exotic phenomena that are often observed in strongly correlated electron materials, such as a metal-insulator transition, a quantum spin-liquid state, and non BCS-type superconductivity. Low-energy properties are described by a molecular orbital, two of which in a dimer form bonding and antibonding orbitals. When the bonding or antibonding band is half filled, the system may be a Mott insulator if interactions between electrons are strong enough. A recent experiment for those compounds reported a dielectric anomaly, indicating the existence of active electric dipole moments, which can be generated by asymmetric distributions of electrons in the dimers. The excitonic insulator (EI) state was first predicted in the middle of last century. The effective attractive interaction between electrons and holes, originating from the repulsive Coulomb interaction, makes an original semiconducting or semimetal state unstable and gives rise to a spontaneous hybridized state of the valence and conduction electrons. Recently, experimental observations of EI states have been reported in chalcogenides and transition metal oxides, e.g., Ta_2NiSe_5 and $LaCoO_3$. Since the EI order parameter generally has the form of an inter-band mixing which is difficult to observe directly in experiments, it is important to explore observable physical quantities characterizing EI states.

In this dissertation, I study the optical responses in the multi-orbital systems with strong electron correlation. We clarify the optical responses at finite temperature in both the long-range ordered and disordered phases. In particular, we take into account the correlation effect, finite temperature effect, the long-range order effect and the multiorbital effect on an equal footing, in contrast to the previous

theoretical researches. I set up the following two detailed themes: 1) To clarify characteristics of the optical responses in the dimer-type organic molecular system with strong electron correlation in both the Mott insulating state and the polar charge ordered phases. 2) To clarify characteristics of the optical responses in the EI system with strong electron correlation and compare with the optical responses in the low-spin band insulator and high-spin Mott insulator.

In order to achieve these purposes, we adopt the extended two-orbital Hubbard models for the dimer-type organic molecular systems and the EI systems. We use one of the most powerful techniques in the quantum cluster method, the variational cluster approach, to consider the local electron-electron correlation exactly and the possible long-range orders, which usually appear only in the thermodynamic limit by the spontaneous symmetry breaking. The optical conductivity spectra are calculated by the linear response theory with vertex correction.

We calculated the finite temperature phase diagram. The results have been compared with our previous studies by the mean-field and Monte-Carlo methods. A competition between the AF phase and the CO phase is found. The one particle excitation spectral have been calculated to study the electron structure. In the strong dimerization regime, we assigned the bonding and antibonding bands. The bonding band is further split by the effective Coulomb interaction U_{eff}. The optical conductivity spectra have been calculated. According to the arrangement of the dimers, a Hubbard peak is found in the σ_{xx}, and a dimer peak is found in the σ_{yy}. The soften model near the CO-DM phase boundary is not fully reproduced due to the small cluster we have adopted. We have shown that the peaks positions are influenced by the appearance of the AF phase in the temperature dependent optical conductivity spectra. With decreasing temperature as the AF phase appears, the Hubbard peak shifts into high energy regime in the DM phase, while the peak energy of the Hubbard peak is not influenced too much in the CO phase. The dimer peak in the CO phase shifts to lower energy regime while it is not influenced too much in the DM phase. The shift of the Hubbard peak is due to the doubling of the Brillouin zone. The shift of the dimer peak is due to the exchange energy induced by the AF.

As for the EI system, we confirmed the spin state transition induced by the competition between Hund's coupling and energy split. We calculated the finite temperature phase diagram and shown the competitions between the LS state, the ESDW state, the HL state, and the HS (AF) state. We calculated the one particle excitation spectra and the optical conductivity spectra. We find in low temperatures that a peak structure appears only in the EI phase. This peak intensity is scaled by the EI order parameter in low temperature. We suggest that this is available to identify the EI phase. At high temperature, we found a similar peak at the same position of EI peak. This peak, however, has a different origin and has been explained as the thermal fluctuation. This point was proved by the benchmark calculation of temperature dependent optical conductivity spectra at the LS phase.

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論文審査の結果の要旨

電子軌道縮退と強い電子間相互作用は、遷移金属化合物、希土類金属化合物、有機固体等の凝縮系において幅広く存在し、その軌道自由度と電子間相互作用の相乗効果により、多極子秩序、非 BCS 的な超伝導、非自明な磁気秩序などの新奇物性が現れると考えられている。これまでの実験・理論研究の多くは、このような系の全容を明らかにする励起状態の重要性を認識させるものであり、特に励起状態を直接解析できる光学スペクトルの観測や数値計算が実験・理論共に盛んに行われてきた。しかしながら、実験と比較可能な理論結果を得るためには、系が有する強相関効果、有限温度効果、軌道自由度の効果、長距離秩序の効果などの様々な効果を考慮した上で、励起状態を適切に取り扱う解析方法を用いる必要があり、このような理論解析を用いた光学スペクトルの計算が強く望まれている。

LI, Hengyue 提出の博士論文は、多軌道強相関電子模型における基底状態と光学スペクトルを理論解析により調べたものである。拡張された 2 軌道ハバード模型を考え、変分クラスター近似法(VCA 法)と呼ばれる解析手法を適用することで、局所的な相関効果、長距離秩序の効果、有限温度効果を同時に適切に取り扱うとともに、光学電気伝導度スペクトルの計算において、ダイアグラム法における頂点補正(vertex correction)の効果を取り入れた定式化を行った。これにより光学スペクトルにおける局所電子相関の効果を取り入れることが可能となり、以下の二つの系についてそれぞれ新しい結果を得た。１）分子二量体を有する分子性固体において、光学スペクトルにおける二つの特徴的なピーク構造のエネルギーが、磁気転移によりシフトすることを見出した。２）励起子絶縁体における光学スペクトルにおいて、励起子凝縮相に特有なピーク構造を見出し、その強度が励起子凝縮の秩序変数と良い対応を示すことを明らかにした。

これらの研究は固体における強相関電子状態に対して新しい知見をもたらし、提出者が自立した研究活動を行うに足る高度の研究能力と学識を有することを示した。よって LI, Hengyue 提出の論文は博士（理学）の学位論文として合格と認める。