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Abstract

Dual Electron-phonon Coupling Model for Giant Photoenhancements of Dielectric Constant and Electronic Conductivity in SrTiO3

Since some 3D type perovkite compounds have been found in the experiments to give gigantic dielectric constant, in contrast to the ordinary dielectrics, its underlying microscopic mechanism has attractted much attention in the field of solid state theory. As is well-known, the ferroelectric modes play an important roll in the dielectric property, transportation and phase transition of these materials. Many efforts were already performed on the connection of this ferroelectric mode with the quartic or sextic anharmonic oscillators. In experiment, gigantic photo-enhancements of the electronic conductivity and the dielectric constant have recently been observed in $SrTiO_3$. It was also pointed out that, this dielectric enhancement remains to exist only under the ultraviolet (UV) illumination, while vanishes as the illumination is turned off. As for the photo-induced electronic conduction in $SrTiO_3$, it is expected to be an alternative mechanism from that of the ordinary field induced one in metallic systems. However, the microscopic origin of these photo-induced phenomena has not yet been clarified theoretically. Thus, this is just the motivation of the present study.

We will give a short introduction to the soft mode theory in the second chapter since our work will be mainly based on this theory. The spatial structure and the electronic property of this 3D perovskite, $SrTiO_3$, are also stated, since we will focus our efforts only on this compound in the present work. Some key points of the previous studies on the fundamental properties of $SrTiO_3$, are summerized as well. In Chapter 3, a detailed illustration is given to the Super-Para-Electric(SPE) large polarons, from the set-up of the theoretic model to the numerical calculations, and ends up with the impurity effect on this polaron. Combining the experimentally observed dielectric and conductive properties with previous theoretical researches on the model for this crystal, we give a further investigation into the model and come up with a new model for the

photo excited state of the crystal. By which, we can adiabatically obtain the corresponding energy surface and find all the metastable states on it. With the investigation of each of such states, we can find its connection with the dielectric and conductive property of this crystal. We also give an investigation into the most stable quasiparticle state for the many electron system within the adiabatic method and give a description to the possible lattice configurations for the photo excited state of the electron and phonon coupling system. Then it is followed by Chapter 4, the applications of this SPE polaron theory in the interpretation of the photo-induced giant dielectric constant and electronic conductivity. We will first discuss one of the fundamental problems for phonons, phonon softening or phonon hardening with the introduction of *e-p* coupling. Then we will apply our SPE large polaron theory to the experimentally observed static dielectric enhancement in $SrTiO_3$, clarifying the microscopic origin of this photo-induced phase transition. In the following, we will give a phenomenological interpretation to the experimentally reported metallic conduction in $SrTiO_3$ by studying the translational property of the polarons. In Chapter 5, we will discuss the relaxation process of the lattice after the photo-excitation. It has already been studied that for polymers, the formation of the polaron or exciton is an ultra-fast process.[61] In experiment, these states could be detected and predicted from the spectra. However, by virtue of the molecular dynamics theory, the information of the lattice configuration as well as the electronic state can be traced so as to compare with the energetic evolution. Then, the lattice relaxation process could be recognized more clearly. We will first give a description to the molecular dynamics theory for the e-p interacting system. Then, we will apply this method to $SrTiO_3$ and give some detailed descriptions about the relaxation process in this crystal. We will show the formation of the SPE large polaron is an ultra-fast process of about several picoseconds, and how the electron and phonon interaction system releases its energy so as to reach its new stable state. The influence of the electron and phonon interaction strength and the size of the electron and phonon coupling system on the relaxation process will be shown. A summary of all the conclusions of these studies is given at Chapter 6.

論文の審査結果の要旨

最近、$SrTiO_3$ や $KTaO_3$ 等の３次元ペロブスカイにおいて、紫外光誘起による「超常誘電性」相転移が発見され、多くの研究者の興味を集めている。この光誘起相転移に関し、現段階までに観測されている事実は、以下の４点である。

ⓐ４eV 付近の紫外光で線形励起すると、これらの物質の準静的誘電率は10^3～10^4程度、巨視的に増大する。

ⓑ酸素の 2p 軌道から、金属カチオン(Ti^{4+}、Ta^{5+})の 3d 軌道へ、電子が光励起され、同時に金属カチオンの周囲の空間反転対称性は、巨視的に消失する。

ⓒ但し、完全な強誘電的長距離秩序が実際に凍結して現れるには到らない。

ⓓ更に、光励起と同時に、高い移動度を有する負の荷電担体が発生し、この物質の電気伝導度は急激に増大する。

これらの状況から、邱宇君は、紫外光で生成した $3d$ 伝導帯電子が、この物質に固有の誘電型ソフト・フォノンと、線形ではなく２次で非線形に結合し、超常誘電性ラージ・ポーラロンを形成する理論モデルを考案した。このラージ・ポーラロンは、電子を内包し、大きな量子揺らぎにより伝搬する強誘電性ドメインと言い換えることもでき、この理論における中心的な概念である。

同君はこの理論モデルの詳細な解析から、２次の電子格子結合と 3d 電子の強い遍歴性が相俟って、前述のⓐ―ⓒの実験事実（誘電率の巨視的増大、空間反転対称性の巨視的破綻、等々）が矛盾無く自然に導かれ、ⓓについてもその理解への有望な指針が得られることを示した。このように、紫外光誘起「超常誘電性」相転移の微視的機構が理論的に解明された意義は極めて大きいと認められる。

これに加えて、同君は前述のラージ・ポーラロンの種々のクラスター状態（バイ・ポーラロン、バイ・ポーラロン分子、等々）の安定性に関する試算、及びラージ・ポーラロン形成の実時間動力学の研究も既に完成させている。

以上により、審査員一同、同君の前述の研究は博士学位論文として十分な水準のものであると判断した。