Density functional theory study of the electronic structure and transport properties of La, V, Nb, and Ta doped $SrTiO_3$

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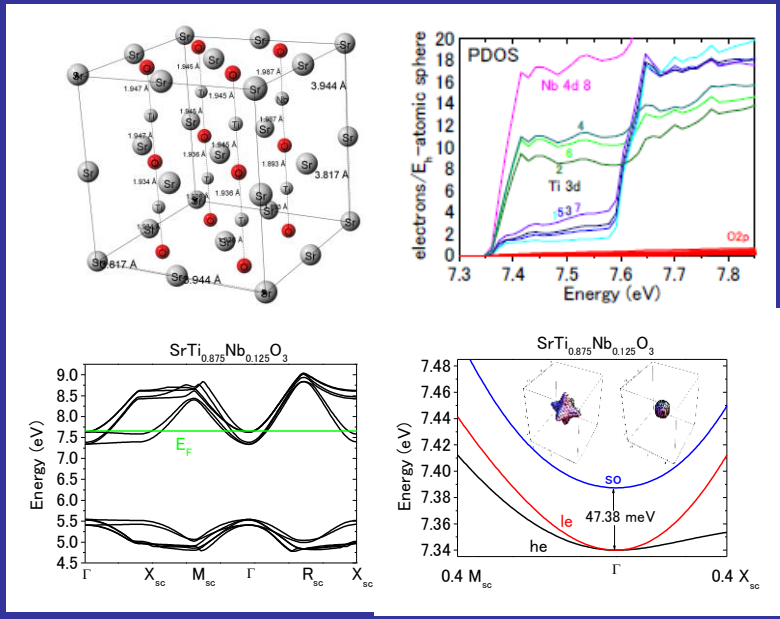
目的: First-principles calculation of crystal structure, electronic structure and thermoelectric properties in donor doped $SrTiO_3$ have been studied in presence of spin-orbit coupling (all calculations used PCC)

内容: Enhancing the electronic transport properties of SrTiO3 (STO) through tuning the electronic structure by dopant substitution is of great interest in view of potential application of STO to thermoelectric energy harvesting devices. A large power factor has been reported for La, Nb and oxygen vacancy doped STO, generating significant interest in theoretical approaches to predict the magnitude, carrier concentration, and temperature dependence of the Seebeck coefficient. In this research, the electronic transport properties of donor doped STO was theoretically studied by first principles calculations including the spin-orbit (SO) interaction and low field Boltzmann transport theory, for pure STO and STO doped with La, V, Nb, and Ta.

結果: The dopant induced changes to the electronic structure, band masses, density-of states masses, split-off energies, and transport properties were systematically studied and the degree to which the calculated Seebeck coefficient (S) predicts the magnitude of the experimental Seebeck data was explored. We have found that, while SO coupling significantly decreases the band masses along the Γ-X direction, the calculated density-of-states masses exhibit a significantly smaller variation for all studied systems, with Seebeck coefficients in the range of -21.39 μV/K to -41.30 μV/K for 1 dopant per 2x2x2 STO supercell. At lower carrier concentrations and temperatures, the SO coupling has a marked effect on both the filling dependence of the density-of-states mass as well as on the temperature dependence of the Seebeck coefficient, with quantitative theoretical predictions of the temperature dependence of S in closer agreement to experimental data than those based on DFT calculations that do not include the SO interaction.

Electronic structure of Nb-doped STO



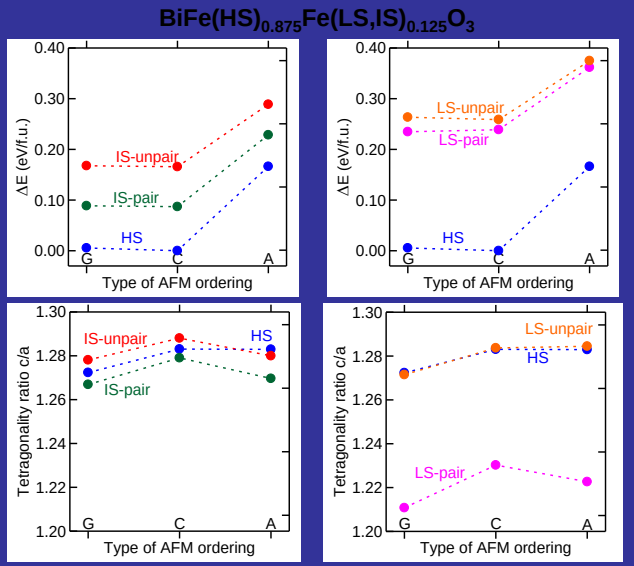
First-Principles Calculation Study of Ferroelectricity and Magnetism in $BiFeO_3$-based materials

東京工業大学総合理工学研究科 Dan Ricinschi

研究目的: First-principles calculation of $BiFeO_3$-based magnetoelectric multiferroics (all calculations used PCC)

研究内容の概要: Magnetoelectric multiferroics attract considerable attention in view of applications to multiple-level non-volatile memories and spintronic devices. In particular, a lot of research is focused on bismuth ferrite ($BiFeO_3$, BFO), due to its rich phase transition portrait including several competing unit cell instabilities. In our research, we use first-principles calculations made under the density functional theory for probing ways to enhance the magnetization of materials from BFO family while preserving robust ferroelectricity and good insulating properties. Previous work has reported pressure-induced phase transitions from high-spin to low-spin or nonmagnetic configuration of Fe^{3+} and Co^{3+} ions, under external pressure. Inspired by the possibility of simultaneously stabilizing colossally-different phases such as a super-tetragonal one with giant c/a ratio and a pseudo-cubic one in BFO, we have systematically analyzed the structure and energetics of supercells with various antiferromagnetic symmetries and electronic configurations of Fe^{3+} ion. We have been particularly interested to analyze the impact of such unconventional spin configurations on the magnetization of the BFO supercell, its tetragonality ratio and ionic displacements as well as spontaneous polarization.

結果: Using the ABINIT first-principles calculation code, we have studied ferrimagnetic arrangements of perovskite blocks inside pure BiFeO3 (BFO) super-cells, by setting various electronic configurations for a part of the Fe^{3+} ions (high-spin, low-spin, intermediate-spin). We have found that some spin configurations of the Fe3+ ion induce dramatic changes of symmetry and atomic displacements in the BFO supercell, while others do not. This allowed us to evaluate the prospected performance of the studied compounds as magneto-electric multiferroics. Specifically, we found that a large spontaneous polarization can be maintained while the magnetization increases in tetragonal BFO with unpaired low-spin and intermediate-spin configurations of Fe^{3+}. Furthermore, tetragonal BFO-based compounds with low-spin and intermediate-spin magnetic ions may allow a large controllable coupling between the spontaneous polarization and the magnetic degrees of freedom.



Quantum Confinement, Electronic Structure, and Thermopower of Perovskite Structure Oxide Heterostructures Explored by Density Functional Theory

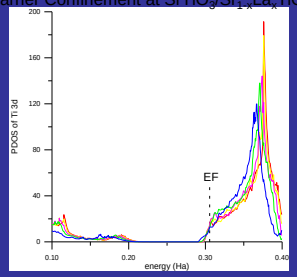
東京工業大学総合理工学研究科 Dan Ricinschi

研究目的: First-principles calculation of $SrTiO_3$ -based quantum well structures (all calculations used PCC)

研究内容の概要: Quantum confinement of carriers utilizing thermoelectric thin films with anisotropic band masses have been predicted to lead to enhancements of the thermopower S of over an order of magnitude compared to bulk. The thermopower is an important consideration for the design of materials systems for efficient thermal to electrical energy conversion as the energy conversion efficiency can be quantified by the figure of merit ZT. In the $SrTiO_3/SrTi_{0.8}Nb_{0.2}O_3/SrTiO_3$ system, large enhancements in S were observed below a $SrTi_{0.8}Nb_{0.2}O_3$ thickness of 16 Å and the origin was attributed to a two-dimensional electron gas confined in the quantum well. In this study, density functional theory is used to explore the electronic structure and thermopower of quantum well structures based on perovskite structure oxide heterostructure systems, with the aim to design advanced materials to increase their thermal to electrical energy conversion efficiency, for use in energy harvesting applications.

結果: Using the ABINIT first-principles calculation code and a theoretical formalism developed based on the energy eigenvalues obtained from DFT, the thermopower of quantum well structures has been studied with differing degrees of confinement length scales, in heterostructures with different conduction band offsets including $SrZrO_3/Sr_{1-x}La_xTiO_3$ and $SrTiO_3/Sr_{1-x}La_xTiO_3$. Moreover, the effect of bulk electronic structures characterized by multifold and single band conduction manifolds has been explored by investigating oxide systems with multifold conduction manifolds resulting from the Ti-3d t_{2g} orbitals ($Sr_{1-x}La_xTiO_3$) and single band conduction manifolds Sn-5s orbitals ($Ba_{1-x}La_xSnO_3$), respectively. The study will resolve the influence of the degree of charge confinement (weak or strong) and the influence of splitting of the bands within a multiband manifold due to confinement on thermopower enhancement in oxide quantum well heterostructures.

Carrier Confinement at $SrTiO_3/Sr_{1-x}La_xTiO_3$ interface



Carrier Confinement at $SrZrO_3/Sr_{1-x}La_xTiO_3$ interface