論文審査の結果の要旨

氏名　侯　秀一

The Doctoral thesis defense for Hou Xiuyi took place on January 16, 2017 at 1 pm in Kashiwa. The defense committee members were professors Terashima, Katsumoto, Matsuda, Matsuura, and Lippmaa.

The topic of the thesis is the analysis of iridium stability in the pulsed laser deposition (PLD) thin film growth process and the growth of IrO_2 and $Pr_2Ir_2O_7$ thin films. The thesis starts with an overview of iridate oxide structures and identifies iridium volatility as a major problem for iridate synthesis in a vacuum process, such as pulsed laser deposition.

Chapter 2 covers the methods and techniques. The main characterization tools for the thin films were atomic force microscopy and x-ray diffraction. Electron microscopy analysis was done with the help of outside collaboration partners. Transport measurements were done by the candidate.

Chapter 3 describes the PLD ablation process and the techniques used for the analysis of the ablation plasma plume. The candidate developed a simple plasma current measurement setup that was used for measuring time-of-flight plasma current profiles for determining the kinetic energy and composition of the ablation plume. The effect of the ablation laser energy density on the kinetic energy of atoms in the plume was measured and the ablation conditions were selected so that resputtering of the film surface by high-energy particles in the plume could be avoided.

Chapter 4 is the main part of the thesis, describing the growth and microstructure of IrO_2 thin films. IrO_2 was selected as a model material for studying the oxidation kinetics of iridium and the volatility of iridium oxides. The main achievement in this part of the work was the mapping of a phase formation diagram for IrO_2 and determining various processes that limit the formation of the IrO_2 phase. A large effort was put by the candidate into analyzing the microstructure, grain formation, and lattice relaxation of the rutile IrO_2 lattice. The necessary scanning transmission electron microscopy work was done in part by prof. T. Yamamoto from Nagoya University. The structural lattice analysis was performed by the candidate.

Chapter 5 contains the second major topic of the thesis -- direct in-vacuum synthesis of $Pr_2Ir_2O_7$ thin films. This material has been predicted to exhibit topological surface states and other exotic electronic phases, but thin films with stoichiometric surfaces have not been made yet. The candidate determined that the biggest obstacle is sputtering-related oxidation and volatilization of iridium, leading to the loss of the pyrochlore phase during thin film growth. A Xe-gas buffering technique was developed to moderate the kinetic energy of the atomic species in the ablation plume. This approach was successful in stabilizing iridium on the surface of the film and, for the first time, the $Pr_2Ir_2O_7$ phase was obtained in a direct physical vapor growth process. Structural analysis of the films showed that phase separation occurs, with PrO_x and Ir metal forming in addition to the pyrochlore phase.

The thesis work made a significant contribution to the field of thin film growth of compounds that contain volatile elements or form volatile oxides. The candidate developed two new techniques that have been rarely applied to PLD growth. One is heavy inert gas buffering that is effective in reducing the kinetic energy of the plume. This technique may potentially help in the synthesis of a variety of new phases that are difficult to grow by conventional PLD. Secondly, a new process monitoring technique was developed based on time-of-flight plume current analysis. This technique is important for monitoring the kinetic energy of the plume and may help in fabricating thin films with dramatically reduced defect densities by PLD due to a better control over film surface sputtering by energetic particles.

The main part of the thesis work, covering the phase formation, grain structure, lattice relaxation, and transport in IrO_2 films has been published in a full paper in the Journal of Crystal Growth.

During the final examination, the referees presented the candidate with many detailed questions regarding the experimental details and the interpretation of the experimental results. The judges identified no major faults in the work, but made several suggestions for further studies. The committee found that the role of the candidate in the thin film process analysis, growth, and characterization experiments was sufficient for granting the doctoral degree.

By unanimous decision, the defense committee agreed that the candidate, Mr. Hou Xiuyi, should be awarded the PhD degree.