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Author(s)	陳, 潔
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博士の専攻分野の名称 博士（理学） 氏名 陳 潔

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High-Pressure Synthesis, Crystal Structures and Physical Properties of Perovskite-Related Mercury and Osmium Oxides

(ペロブスカイトに関連する水銀およびオスミウム酸化物の高圧合成と結晶構造と物性)

Transition metal oxides with partially filled $3d$ and $4d$ shells have dominated materials research in past decades, while $5d$ oxides are being of great current interest as $5d$ oxides exhibit spectacular phenomena. The unique properties of $5d$ oxides stem from the nature of $5d$ electrons themselves. On one hand, upon descending the periodic table from the $3d$ to $5d$ series, the d orbitals become spatially extended, resulting in smaller on-site Coulomb repulsion for $5d$ electrons. On the other hand, the spin-orbit coupling increases considerably in $5d$ orbital, leading to enhanced splitting degenerate orbitals and bands. The interplay between spin-orbit coupling, Coulomb repulsion and crystal-electric field can lead to novel phases of matter. In this thesis, eight $5d$ oxides were synthesized under high-pressure and high-temperature conditions to elucidate the novel physical properties involving $5d$ oxides.

Chapter 1 is the introduction of background behind this thesis. Chapter 2 provides information about the experimental methods and technologies used in this thesis.

In Chapter 3, HgPbO_3 is introduced, which was synthesized at 6 GPa and its crystal structure was studied by single-crystal X-ray diffraction and powder synchrotron X-ray diffraction measurements. HgPbO_3 crystallizes into a hexagonal structure ($R\bar{3}m$) with $a = 5.74413(6)$ Å and $c = 7.25464(8)$ Å. The metallic behavior was observed from temperature dependence of resistivity measurement for HgPbO_3 , but its weak temperature dependence of the electrical resistivity, significant diamagnetism, and remarkably small Sommerfeld coefficient imply that HgPbO_3 is a semimetal. The discovery of semimetal in oxide compounds is quite rare,

which could be useful for the advanced material research and the development of multifunctional devices.

Chapter 4 summarizes two new osmium triple perovskite oxides synthesized at 6 GPa and 1100 °C; a polymorph of the triple perovskite $\text{Ba}_3\text{CuOs}_2\text{O}_9$, which exists in the orthorhombic phase ($Cmcm$), is reported. The crystal structure of $\text{Ba}_3\text{CuOs}_2\text{O}_9$ transfers to a hexagonal structure ($P6_3/mmc$) under a high-pressure and high-temperature condition. Although $\text{Ba}_3\text{CuOs}_2\text{O}_9$ maintains the 6H perovskite-type lattice basis, the distribution of Cu and Os atoms are dramatically altered over the lattice. The new distribution brings about distinct magnetic properties; for example, the hexagonal $\text{Ba}_3\text{CuOs}_2\text{O}_9$ exhibits a ferrimagnetic transition at 290 K in stark contrast to the antiferromagnetic transition at 47 K exhibited by the orthorhombic $\text{Ba}_3\text{CuOs}_2\text{O}_9$. Furthermore, it was achieved by replacement of Cu by Ni that the T_c was successfully increased to 370 K for $\text{Ba}_3\text{NiOs}_2\text{O}_9$. In this chapter, a new class of 3d-5d hybrid ferrimagnetic materials with high- T_c emerged from the application of high-pressure technique. $\text{Ba}_3\text{CuOs}_2\text{O}_9$ as well as $\text{Ba}_3\text{NiOs}_2\text{O}_9$ could serve as a worthwhile platform for developing high- T_c ferrimagnetic material.

In Chapter 5, the study of substitutional series double perovskite oxides $\text{Sr}_2\text{Cr}_{1-x}\text{Ni}_x\text{OsO}_6$ ($x = 0, 0.25, 0.5, 0.75$ and 1) prepared at 6 GPa is presented. $\text{Sr}_2\text{Cr}_{1-x}\text{Ni}_x\text{OsO}_6$ ($x = 0, 0.25, 0.5, 0.75$ and 1) exhibit structural transition from cubic to tetragonal at $x = 0.5$. The former three samples ($x = 0, 0.25$ and 0.5) exhibit ferrimagnetic ordering below T_c , while the other two Ni-rich samples ($x = 0.75$ and 1) are antiferromagnetic. Remarkable variation of the magnetic properties among the five samples seems not to be simply explained by the structural transition, but rather by 5d electron nature. The series of Ni-substituted $\text{Sr}_2\text{CrOsO}_6$ likely gives an insight into the effect of spin-orbital coupling on the novel physical properties.

In Chapter 6, the general conclusion is provided, and prospects for future research of 5d oxide are presented.