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授与した学位	博 士
専攻分野の名称	理 学
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学位授与の要件	自然科学研究科 地球生命物質科学専攻 (学位規則第4条第1項該当)
学位論文の題目	Study on preparation of new superconducting materials and their detailed characterization in a wide pressure range (新しい超伝導物質の作製と広い圧力領域での詳細な特性評価に関する研究)
論文審査委員	教授 久保園 芳博 准教授 後藤 秀徳 教授 横谷 尚睦
学位論文内容の要旨	
<p>Superconductivity of titanium-based pnictide oxides, $\text{Ba}_{1-x}\text{A}_x\text{Ti}_2\text{Sb}_2\text{O}$ (A: Na, K and Rb) and $\text{BaTi}_2(\text{Sb}_{1-y}\text{Bi}_y)_2\text{O}$, has been discovered during the past decade. Here, the superconductivity emerged through the suppression of the charge density wave (CDW) / spin density wave (SDW) transition. From the above results, the titanium-based pnictide oxide superconductors may provide a new platform to study and discuss the correlation between superconductivity and nonmagnetic / magnetic ordered state. In this doctoral thesis, the titanium-based pnictide oxide superconductors were prepared, and their superconducting properties were fully investigated in a wide pressure range. On the other hand, the topological materials such as topological insulators and Weyl / Dirac semimetals have been extensively studied for the expectation not only to new physics but also to diverse applications. In this doctoral thesis, the superconducting properties of possible Dirac semimetals, $\text{Pt}_{1-x}\text{Pd}_x\text{Te}_2$ and BaBi_3, were investigated owing to much interest in topologically nontrivial nature in superconductivity.</p> <p>In chapter 3, the preparation of titanium-based pnictide oxide compounds, $\text{Ba}_{1-x}\text{A}_x\text{Ti}_2\text{Sb}_2\text{O}$ (A: K and Rb), and their superconducting properties are reported, demonstrating that these materials are type-II superconductors. The temperature (T) dependence of electrical resistance (R) was measured at 0 – 3.09 GPa. The superconducting transition temperature, T_c, of $\text{Ba}_{1-x}\text{Rb}_x\text{Ti}_2\text{Sb}_2\text{O}$ decreased with an increase in pressure up to 3.09 GPa. In chapter 4, the preparation of new superconducting titanium-based pnictide oxide compound, $\text{Ba}_{1-x}\text{Cs}_x\text{Ti}_2\text{Sb}_2\text{O}$, is reported, and its superconducting properties are demonstrated in a wide pressure range. Very interesting T_c – pressure (p) behavior was observed. The value of T_c decreased with an increase in pressure up to 4.0 GPa, but it increased above 4.0 GPa, indicating that it may not be a simple BCS type superconductor. No structural phase transitions were observed at 0 – 23.4 GPa. Therefore, the above T_c – p behavior may be related to the electronic transition such as Lifshitz transition. In chapter 5, the preparation of superconducting $\text{BaTi}_2\text{Bi}_2\text{O}$ is reported. The value of T_c was 4.33 K. The crystal structure and superconducting properties of $\text{BaTi}_2\text{Bi}_2\text{O}$ are demonstrated in a wide pressure range. The R – T plots were recorded at different pressures to determine the value of T_c. The value of T_c increased monotonously with an increase in pressure up to 4.0 GPa, and it saturated above 4.0 GPa. The reduced critical field (h^*) – normalized temperature ($t (= T / T_c)$) plot at 7.28 GPa suggested the deviation from the simple s-wave dirty limit model, suggesting the unconventional nature in superconductivity. In chapter 6, the preparation and characterization of a possible type-II Dirac semimetal, $\text{Pt}_{1-x}\text{Pd}_x\text{Te}_2$ ($x = 0.25$), are demonstrated. The value of T_c was ~3.2 K at ambient pressure. The value of T_c was almost constant at 0 – 8.18 GPa. Interestingly, the high T_c value of 7.2 K was recorded above 4 GPa for one sample of $\text{Pt}_{1-x}\text{Pd}_x\text{Te}_2$ prepared in this study. The exact characterization of 7.2 K superconducting phase may be important. In chapter 7, the author reports preparation of superconducting BaBi_3, and its superconducting properties are demonstrated in a wide pressure range. The unconventional superconductivity was suggested from $h^* - t$ plot at ambient pressure. The $T_c - p$ plot of BaBi_3 was fully investigated up to 10.5 GPa, and the value of T_c was almost constant against pressure at 0 – 10.5 GPa. Therefore, applying more pressure for BaBi_3 may be significant to explore the electronic transitions.</p> <p>Thus, the author discovered unconventional superconducting properties in titanium-based pnictide oxides and possible Dirac semimetals. Some interesting $T_c - p$ behavior found in the above materials could not be associated with the structural variation against pressure. Therefore, it may be correlated with the electronic transitions. Also, Cooper pair symmetry for some superconducting phases may not be ascribable to the simple s-wave dirty limit mode, suggestive of unconventional superconductivity. The origin of unconventional superconductivity remains to be clarified, but through this study, the author clearly demonstrated good target materials for studying unconventional superconductivity as well as the strange $T_c - p$ behavior which cannot be explained within the framework of a simple BCS theory.</p>	

論文審査結果の要旨

WANG Yanan 氏の学位論文は、 $\text{BaTi}_2\text{Sb}_2\text{O}$ のBaをCsによって置換した物質の超伝導体合成と圧力下での超伝導特性の研究、ならびに $\text{BaTi}_2\text{Bi}_2\text{O}$ の超伝導特性を広範な圧力領域で詳細に研究した結果を最初の報告としている。前者の物質では、電荷密度波（CDW）もしくはスピン密度波（SDW）に帰属される相転移を抑える中から超伝導が出現しており、一部CDW/SDW秩序状態と超伝導が共存するという興味深い特性が見いだされる。WANG氏は、新規な物質としての $\text{Ba}_{1-x}\text{Cs}_x\text{Ti}_2\text{Sb}_2\text{O}$ を合成するとともに、超伝導転移温度の圧力依存性を調べ、超伝導転移温度が3.7 GPa以上で上昇を開始するという興味深い特性を明らかにした。同氏は、圧力下でのX線回折パターン測定によって、圧力下で構造相転移が起こっていないことを確認した上で、この起源をLifshitz転移のような電子転移が起こっているためと結論付けた。 $\text{BaTi}_2\text{Bi}_2\text{O}$ は、最近Dirac半金属であることが示唆された新規なトポロジカル量子物質である。同氏は、高圧での超伝導転移温度の磁場依存性から、ペアリングの対称性を追求し、 p 波超伝導の可能性を示唆した。これによって、Dirac半金属がトポロジカル超伝導体になりうることを主張した。更に、超伝導転移温度が圧力印加とともに上昇し、4 GPa以上で一定になるという興味深い特性を見いだした。

WANG氏は、次にDirac半金属である $\text{Pt}_{1-x}\text{Pd}_x\text{Te}_2$ ならびに BaBi_3 の超伝導特性を広範な圧力領域において研究して、 $\text{Pt}_{1-x}\text{Pd}_x\text{Te}_2$ においては超伝導転移温度が7 Kとなる新規な超伝導相が圧力下で出現することを示した。一方、 BaBi_3 については、常圧での超伝導転移温度の磁場依存性から p 波超伝導であることを示唆する結果を得て、Dirac半金属の超伝導特性がトポロジカルに非自明であることを例証した。また、これまで行われてきたよりも広い圧力領域での超伝導特性を解明した。これらの研究は、圧力下での超伝導特性研究をDirac半金属において実施した稀有の例である。とくに、Dirac半金属と超伝導特性の相関を明らかにしようとする野心的な試みは、超伝導物理学ならびに超伝導化学上の重要な研究課題であり、博士の学位を付与するにふさわしいものである。