学位論文の要旨
Abstract of Thesis

研究科 School	自然科学
専　攻 Division	地球生命物質科学
学生番号 Student No.	51428252
氏　名 Name	杨　小帆

学位論文題目 Title of Thesis（学位論文題目が英語の場合は和訳を付記）

Study on superconductivity of various types of new superconductors under pressure
(圧力下での様々な種類の超伝導体の超伝導性に関する研究)

学位論文の要旨 Abstract of Thesis

Pressure application is one of effective approaches for tuning and controlling physical properties of materials, with providing the exotic materials and phases which have never been realized at ambient pressure. Various effects of application of pressure on materials, such as a decrease in unit cell volume, a structural phase transition, and a modification of electronic states, have been extensively investigated and employed for new materials design in solid state physics and chemistry. Especially, an application of pressure is an efficient approach for tuning the superconducting transition temperatures, T_c's, of superconducting materials without any chemical modification. The approach may lead to the discovery of exotic superconducting materials, as well as the realization of high-T_c superconductors. Actually, a pressure-driven superconducting phase was successfully discovered in various materials [1-7]. The recent topic is high-T_c superconductivity found in hydrides, H_2S and LaH_{10}, under extremely high pressure [6,7]. Consequently, a pursuit of superconducting phases by applying pressure for materials must be very attractive from viewpoint of both chemistry and physics.

Throughout this doctor thesis, the author reports the fascinating superconducting properties under pressure in recent topical superconductors, and discusses the origin of the superconducting behavior observed under pressure. This doctor thesis includes seven chapters. In chapter 1, the author introduces various techniques for pressure experiments, as well as outline of superconductivity and pressure-induced superconductivity. Namely, the background of this doctor thesis is described in chapter 1. In chapter 2, the purpose and motivation of this study are fully described. The author aims at searching for new superconducting phases under pressure, in particular the phases with exotic superconducting behavior and high-T_c value by suitably employing the techniques of pressure application.

Name	杨　小帆

In chapter 3, the superconducting behavior of a new binary-elements graphite, $Ca_xCs_{1-x}C_y$, is reported under pressure. The $Ca_xCs_{1-x}C_y$ takes a CsC_8-type crystal structure (hexagonal structure, space group of No. 181, $P6_422$) [8], which was prepared by an intercalation of Ca and Cs in highly oriented pyrolytic graphite *via* the metal-alloy method. The T_c – pressure (p) plot showed a dome shaped behavior without any structural phase transitions in a wide pressure range, *i.e.*, a clear positive pressure effect on T_c was obtained below 9.3 GPa, while the T_c value rapidly decreased with a further increase in pressure. The T_c value reached 11.7 K at 9.3 GPa. The positive pressure effect is probably caused by the pressure-driven softening of in-plane Ca(Cs)-Ca(Cs) phonons, and the subsequent negative pressure effect was attributed to the order-disorder transition (or random off-center displacement of Ca(Cs) atoms), as was indicated in CaC_6 [9].

In chapter 4, the pressure dependence of superconductivity in a new topical iridate (Ir) superconductor, $SrIr_2$, is fully reported, in which Ir is a 5*d* transition metal with a strong spin orbit coupling (SOC). The T_c value of $SrIr_2$ was as high as 6.6 K at ambient pressure. The T_c value monotonously decreased with an increase in pressure, implying a negative pressure effect on T_c. On the other hand, the onset superconducting transition temperature, T_c^{onset}, increased above ~8 GPa, indicating an upward turn of T_c^{onset} in a high pressure range. This upward turn was reproduced in multiple samples of $SrIr_2$. The pressure-dependent X-ray diffraction suggested no structural phase transitions in a pressure range of 0 – 22.4 GPa. Therefore, the upward turn of T_c^{onset} was not attributed to the structural phase transition but presumably electronic transition. The magnetic field, H, dependence of T_c recorded under pressure for $SrIr_2$ suggested that the paring was explained by neither *s*-wave clean nor dirty limit superconductivity (or simple *s*-wave BCS-type conventional superconductivity), which may be associated with the anomalous T_c^{onset} - p plot observed for $SrIr_2$.

In chapter 5, the author reports the pressure dependence of electrical transport in a new type of superconducting iridium silicide compound, Li_2IrSi_2, in order to clarify the superconducting behavior under pressure. An anomalous behavior of superconductivity against pressure was strongly expected for Li_2IrSi_2 because of exotic physical properties expected from the strong SOC of Ir. However, only a negative pressure effect on T_c was observed, *i.e.*, the T_c value monotonically decreased with increasing pressure up to 7.03 GPa, and it disappeared above 7.03 GPa. The temperature dependence of electrical resistance (R) substantially showed only a metallic behavior at normal state in all pressure range of 0.790 – 16.4 GPa. This study is not yet completed, but it may be the first step for clarification of superconducting behavior of Li_2IrSi_2 under pressure.

In chapter 6, the author reports the superconducting behavior of $(NH_3)_yLi_xFeSe_{0.5}Te_{0.5}$ under pressure. Two superconducting phases of $(NH_3)_yLi_xFeSe_{0.5}Te_{0.5}$ were realized, which showed the T_c values as high as 20.2 and 29.5 K at ambient pressure, called as 'low-T_c phase' and 'high-T_c phase'. The R – T plot was recorded for low-T_c phase of $(NH_3)_yLi_xFeSe_{0.5}Te_{0.5}$ over a pressure range of 0 –

Name	杨　小帆

14 GPa, and for high-T_c phase of $(NH_3)_yLi_xFeSe_{0.5}Te_{0.5}$ over 0 – 19 GPa, yielding double-dome superconducting $T_c – p$ phase diagrams *i.e*., two superconducting phases (SC-I and SC-II) were found for both the low-T_c and high-T_c phases under pressure. For the low-T_c phase, the maximum T_c was 20.0 K at 0 GPa for SC-I, and 19.9 K at 8.98 GPa for SC-II. For the high T_c-phase, the maximum T_c was 33.0 K at 1.00 GPa for SC-I, and 24.0 K at 11.5 – 13.2 GPa for SC-II. These results imply that the maximum T_c value of high pressure phase (SC-II) does not exceed the maximum value of SC-I, unlike what was shown in the $T_c – p$ phase diagrams of $(NH_3)_yLi_xFeSe$ [10] and $(NH_3)_yCs_xFeSe$ [11] investigated previously. Moreover, any structural phase transition was not observed for either the low-T_c or high-T_c phase of $(NH_3)_yLi_xFeSe_{0.5}Te_{0.5}$ over the wide pressure range of 0 to 15.3 GPa. The T_c – lattice constant (*c*) plots for both phases were recorded to determine the critical point separating SC-I and SC-II.

In chapter 7, the author summarizes the results shown in chapters 3 – 6. Throughout the study of this doctor thesis, the superconducting behavior for three types of topical materials is fully investigated under pressure. The $R – T$ plots and X-ray diffractions were measured in a wide pressure range, in order to clarify the superconducting behavior and crystal structures for the materials. In particular, the exotic superconducting behavior under pressure was pursued for the recent topical materials. Consequently, the positive pressure dependence of T_c which is not simply explained by the concept of conventional superconductor was discovered in some of target materials. This is very significant from view of not only pioneering new superconductor physics (or development of new exotic superconductors), but realizing high-T_c superconductors. This study would become the significant step for generating new exotic / high-T_c superconductors by suitably employing pressure.

References

[1] T. He *et al*. Phys. Rev. B **97**, 104503 (2018).
[2] T. He *et al*. Phys. Rev. B **100**, 094525 (2019).
[3] T. T. M. Palstra *et al.* Solid State Commun. **93**, 327 (1995).
[4] T. Imai *et al.* Phys. Rev. Lett. **102**, 177005 (2009).
[5] L. Sun *et al*. Nature **483**, 67 (2012).
[6] A. P. Drozdov *et al*. Nature **525**, 73 (2015).
[7] M. Somayazulu *et al*. Phys. Rev. Lett. **122**, 027001 (2019).
[8] D. Guerard *et al*. Carbon **16**, 285 (1978).
[9] A. Gauzzi *et al*. Phys. Rev. Lett. **98**, 067002 (2007).
[10] P. Shahi *et al*. Phys. Rev. B **97**, 020508(R) (2018).
[11] M. Izumi *et al*. Sci. Rep. **5**, 9477 (2015).