論文の内容の要旨

Electronic states of narrow-gap semiconductors under multi-extreme conditions
（多重極限環境下における ナローギャップ半導体の電子状態）

氏名：秋葉　和人

Materials can be classified into metals or insulators from the viewpoint of the electric conduction. Exploration of exotic electronic phases beyond the conventional framework of the band theory has been a challenging subject in condensed matter physics.

More than 50 years ago, emergence of the excitonic insulator phase, a quantum condensed state of electron–hole pairs, was theoretically proposed as a new ground state at the boundary between metal and insulator. Although numbers of experimental attempts have been devoted to search the realization of the excitonic insulator phase, the concrete evidence have never been reported up to the present.

The boundary region between metal and insulator comes to attract renewed interests in viewpoints of topology in condensed matter physics. The topological materials are characterized by their non-trivial band topology and high mobility carriers obeying the relativistic Dirac equation. Since Dirac equation originally describes the motion of particles with high velocity close to the light speed, transport properties of topological materials attract general interest beyond the field of condensed matter physics. Actually, numbers of unconventional behaviors have been reported experimentally, yet the essence stemming from the non-trivial topology of the band structure remains unclear since the interpretation of experimental results is often hindered by their complex electronic structures.

Common to these topics, model materials having simple and controllable band structure are needed. As candidates for such materials, two narrow-gap semiconductors, black phosphorus (BP) and lead telluride (PbTe), were investigated in this thesis.

We utilized static magnetic fields up to 14 T and pulsed magnetic fields up to 55 T. The electrical transport measurements under pressure were performed in static fields with piston-cylinder-type pressure cell. Resistivity, magnetization, and ultrasonic measurement techniques

under pulsed high magnetic fields were utilized in this study.

BP is known as a high mobility semiconductor with narrow direct band gap. The band gap is suppressed by applying hydrostatic pressure, and is expected to collapse at a certain pressure. This suggests that the band structure can be tuned continuously from semiconductor to semimetal, which is an ideal playground to explore the exotic states in the vicinity of the semiconductor-semimetal (SC-SM) transiton. Since only carriers near the narrowest band gap govern the physical properties, BP under pressure is regarded as a quite simple playground. Although several previous studies demonstrated the metallization under pressure, the details of the electronic states around the SC-SM transition was unclear. Thus, we intend to clarify the electronic structure in the vicinity of the SC-SM transition.

We investigated the electrical transport properties in pressurized BP under high magnetic fields. In semiconducting states below 1 GPa, we observed the magneto-phonon resonance (MPR), which is a quantum transport phenomenon showing up in high mobility semiconductors. Through the analysis of the MPR, we identified a reduction of the cyclotron mass with applying pressure, which is understood as a result of the band gap reduction. At higher pressures, we observed clear Shubnikov-de Haas (SdH) oscillations, which directly demonstrate the pressure-induced SC-SM transiton. We revealed the light cyclotron masses and small carrier densities in the semimetallic state, which are comparable with conventional elemental semimetals, bismuth and graphite. Further, the Fermi surface of semimetallic BP becomes monotonically larger as pressure increases, which indicates the tunability of carrier density by hydrostatic pressure. The origin of large and non-saturating magnetoresistance in semimetallic BP was discussed by the semiclassical two-carrier model. We found that the magnetoresistance cannot be fully reproduced by the Drude model. Therefore, additional mechanisms such as change in carrier relaxation time in magnetic fields have to be considered. In addition, the nearly compensated nature and large difference of the mobilities between electrons and holes were clarified from the two-carrier analyses. To explore the exotic electronic states near the SC-SM transition, we measured the temperature dependence of the resistivity down to 43 mK in magnetic fields. Although apparent semimetal-to-insulator-like change was observed in the temperature dependence of the longitudinal and transverse magnetoresistance, these phenomena were reasonably reproduced as trivial effects within the conventional theory for metals in high magnetic fields.

PbTe is known as a degenerate direct-gap semiconductor, which has moderate carrier density of $\sim 10^{18}$ cm^{-3} at low temperature. Substitution of Sn for Pb and application of hydrostatic pressure reduce the band gap, and cause the topological phase transition to the topological crystalline insulator (TCI). Recent theoretical study suggested that the ratio of Zeeman energy to cyclotron energy (ZC ratio) can be used as a quantitative index to determine the degree of similarity to the Dirac electron system, namely, "Diracness" of materials. ZC ratio is known to be unity in a system with ideal two-band Dirac Hamiltonian. Theoretical calculation expected that the ZC ratio increases by substitution of Sn, and becomes unity at the zero-gap state, namely, the topological phase transition point. This suggests the ralization of ideal Dirac electron system in this material. PbTe is suitable to testify above new criterion to identify the "Diracness" for its simple band structure and strong spin-orbit effect. Besides, we can seek the unconventional behavior by tuning the band topology from a trivial semiconductor to TCI *via* the zero-gap state by manipulating external parameters. Based on motivations mentioned above, we investigated various physical properties of PbTe and $\mathrm{Pb}_{1-x}\mathrm{Sn}_x\mathrm{Te}$ in magnetic fields.

We studied the electrical transport, magnetization, and elastic properties in PbTe, and observed clear SdH, de Haas-van Alphen (dHvA), and acoustic dHvA oscillations. We explained that the large second harmonic observed in the quantum oscillations is due to the prominent spin-splitting in PbTe. We analyzed the spin-splitted quantum oscillation based on the conventional Lifshitz-Kosevich formula, and found the oscillation pattern was reproduced by the ZC ratio. The simple band structure of PbTe and high-field measurements up to 55 T enabled us to unambiguously determine the ZC ratio of 0.52. From these results, we clarified that PbTe is in the spin-polarized quantum limit state above ~35 T. We also observed large and non-saturating magnetoresistance effect in both transverse and longitudinal configurations, which origin was remained as an open question. In transport measurements under pressure, we observed slight upturn of the ZC ratio in one sample, which indicates that the Diracness of PbTe is enhanced by pressure.