1. Investigation of thermoelectric and magneto-transport properties of polycrystalline Pt_5P_2

2. Exploration of superconductivity in high-pressure Laves phase crystals

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Abstract

1. In this work, we have focused on the phosphorus compound Pt_5P_2 and investigated its physical properties from thermoelectric measurements, transport measurements, and DFT calculations. The crystal structure of Pt_5P_2 was first reported in 1967, but its physical properties have not been investigated in detail. In this study, we have established a method for the synthesis of high-purity Pt_5P_2 polycrystals and have also succeeded in obtaining samples with controlled crystallinity. Magnetotransport measurements on the obtained samples revealed a huge positive magnetoresistance of ~10^3% in spite of the polycrystalline nature. Furthermore, the magnetoresistance values show a systematic dependence on the grain size.

2. In this work, we worked on synthesizing the Laves phase crystal, which is an intermetallic compound phase, under high pressure to search for novel superconductors. The powder X-ray diffraction patterns of the synthesized compound could not be indexed to the cubic phase, which is the ambient pressure phase, indicating a structural transition to the high-pressure phase. However, the crystallinity was apparently poor and no superconductivity was observed as far as magnetization measurements are concerned.

In this report, we mainly focus on the results of the first project.

Authors

Alex Hiro Mayo:

He belongs to Kawasaki Laboratory, Department of Applied Physics, the University of Tokyo. He has been working in Ishiwata Laboratory, Graduate School of Engineering Science, Osaka University (which belonged to the University of Tokyo until 2018) for material exploration and magnetotransport measurements of novel magnetic topological semimetals, using techniques such as high-pressure synthesis. In the first project, he worked on thermoelectric measurements, magnetoresistance measurements in the low-field region and electronic structure calculations. In the second project, he worked on high-pressure synthesis and magnetization measurements.

Karolina Górnicka:

She is a member of Prof. Tomasz Klimczuk's group at the Gdańsk Univ. of Technology, Poland. She has been exploring new materials using a wide range of synthetic methods, including solid-state reaction, flux and chemical transport methods. She was in charge of the synthesis of Pt_5P_2 polycrystalline samples for the first project, and also conceived the second project.

Research Background and Objectives

1. Elemental phosphorus is known to have many allotropes by adopting a wide variety of covalent frameworks ranging from molecular to three-dimensional crystal structures [1]. Among these, black phosphorus has long been studied as a narrow-gap semiconductor in which *p*-electrons with high mobility are responsible for conduction, and it was recently reported that it shows a topological transition to Dirac semimetal upon applied pressure [2]. Furthermore, Mayo et al. has recently reported magnetic-field-induced topology switching between magnetic Weyl and nodal-line semimetal phases in α-EuP_3, a material which can be understood as a “magnetic variant” of black phosphorus, incorporating magnetism between the conducting phosphorus layers [3]. Therefore, phosphorus compounds deserve attention as a platform for the development of various crystal and electronic structures and transport phenomena. The crystal structure of Pt_5P_2, which is the focus of this study, was reported in 1967 [4], and it is known to have a honeycomb-like slightly buckled P-covalent network along with Pt octahedra (Fig. 1). However, it is not easy to obtain high-purity samples, and the physical properties such as electronic structure and transport properties have not been explored so far. In the present work, we have investigated the electrical properties of Pt_5P_2 by means of thermoelectric and magnetoresistance measurements of transport properties using high purity Pt_5P_2 polycrystalline samples, and DFT calculations of the electronic structure.

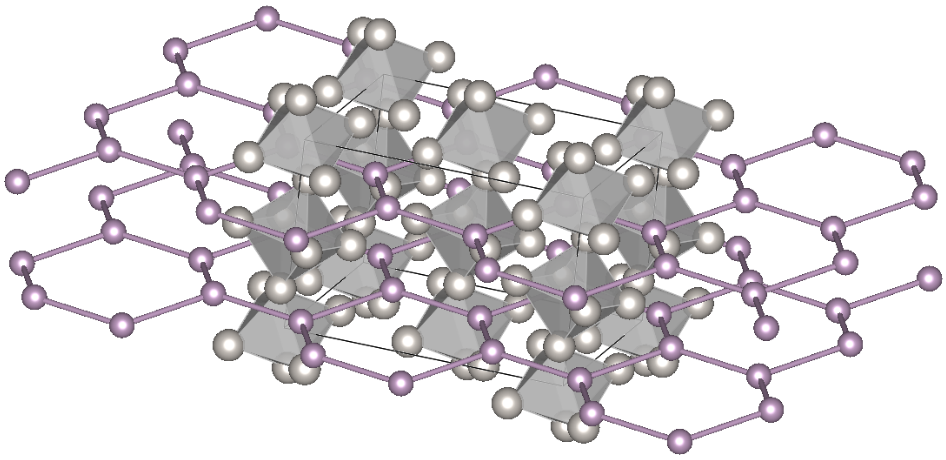

Fig. 1. Crystal structure of Pt_5P_2

2. The Laves phase, represented by AB_2, is one of the most abundant intermetallic phases, with more than 1000 different compounds, including many superconductors. The Laves phase has three different crystal structures: a fcc *C*15-type cubic (*Fd-3m*), a *C*14-type hexagonal ($P6_3/mmc$) and a *C*36-type hexagonal ($P6_3/mmc$), and depending on the pressure conditions, a structural phase transition is expected to occur between these structures. In this research project, we synthesized the material under high pressure, and also explored the superconductivity of the material (the specific name of the target material is not given because it is still unpublished).

Experimental method

Pt_5P_2 polycrystalline samples were synthesized by melting or solid-phase reactions of the ingredients weighed according to stoichiometric ratios. Besides these different reaction methods, the crystallinity was controlled by changing the purity of the Pt powder. The magnetotransport properties were measured by the four-terminal method using the Electrical Transport Option (ETO) of MPMS3 manufactured by Quantum Design. The Seebeck coefficient was measured by the steady-state method using the ETO of MPMS3 and a self-made probe. Electronic structure calculations were performed using the ab initio packages Quantum ESPRESSO [5] and FermiSurfer [6].

The Laves phase intermetallic compounds were synthesized by high-pressure synthesis method and their structures were evaluated by powder X-ray diffraction. Magnetization measurements were also carried out using MPMS3.

Results and Discussion

1. The temperature dependence of the magnetoresistance and Seebeck coefficient of the Pt_5P_2 polycrystalline sample with the largest grain size among the synthesized samples is shown in Fig. 2. In general, the magnetoresistance of polycrystalline materials is expected to be small due to the effect of grain boundary scattering, but in this study, a huge positive magnetoresistance of exceeding 6000 % was observed under a magnetic field of 7 T. The Seebeck coefficient measured for the same sample is small in absolute value and shows a sign reversal during the cooling process, which is characteristic of a semimetal with coexisting electrons and holes.

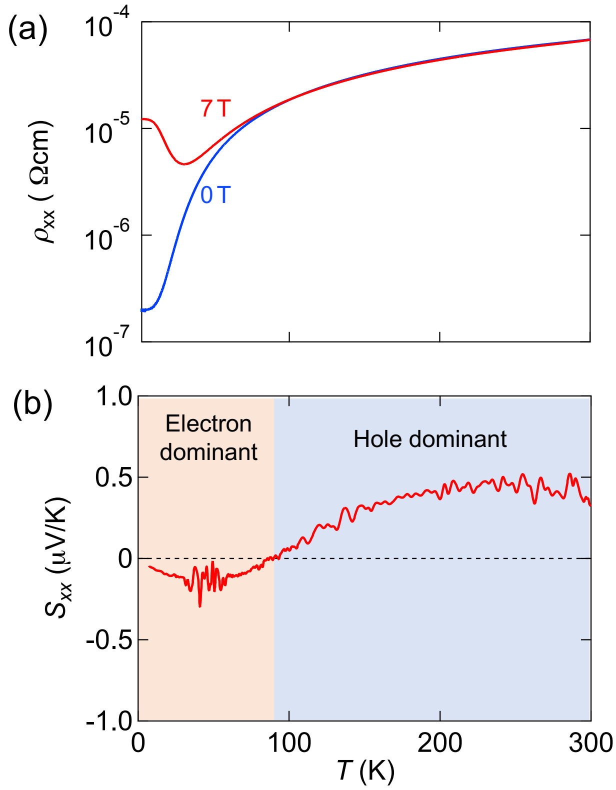

Fig. 2. Temperature dependences of the (a) resistivity and (b) Seebeck coefficient, respectively.

Figures 3(a) and (b) show the results of the electronic structure calculations using the parameters obtained from the synthetic sample, which show the coexistence of electron and hole pockets as expected from Fig. 2. In addition, the Fermi surface shows "open orbitals" which are not closed in the Brillouin zone, both of which are considered to contribute to the huge magnetoresistance. Finally, the sample dependence of the magnetotransport properties is shown in Fig. 3(c). The data for other materials are based on [7]. The magnetoresistance of the five samples was found to increase systematically with increasing crystallinity. One possible explanation for this behavior is the systematic change in mobility, i.e., in ideal semimetals, the magnetoresistance tends to increase in proportion to the product of the electron and hole mobilities. Since the magnetoresistance in this study is proportional to the low-T conductivity to the power of 1.7, it is possible that the present case is relatively close to this picture. Another possibility is the realization of ballistic conduction, in which the mean free path of the conduction carriers is as large as the grain size.

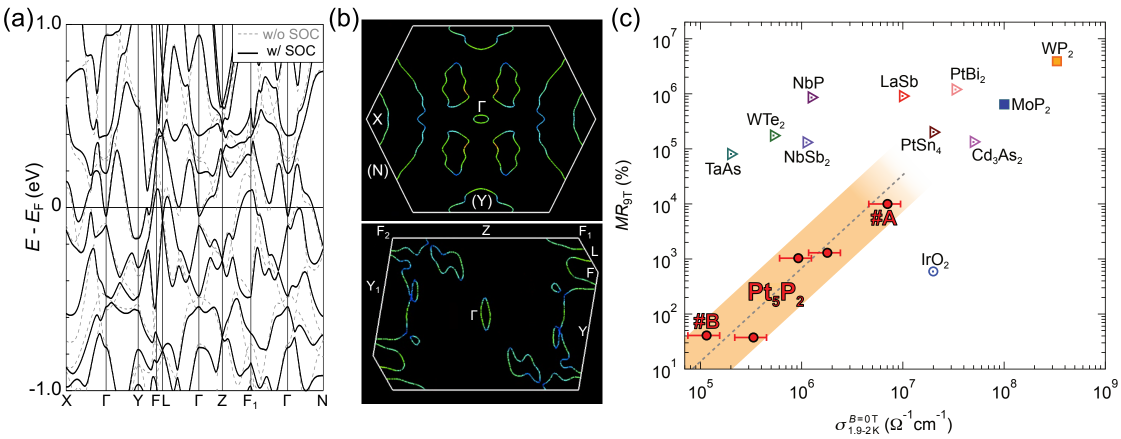

Fig. 3. (a) Band structure and (b) Cross sections of the Fermi surface.
(c) MR comparison among reported materials

2. As for the Laves phase intermetallic compound, powder X-ray structure diffraction after high-pressure synthesis showed a diffraction pattern with broad peaks. This result suggests that the crystallinity is poor and the peaks could not be assigned. The obtained sample was used for magnetization measurements, and no superconductivity was observed in the temperature range up to 1.8 K, the limit of measurement.

Future Outlook

In addition to the huge positive magnetoresistance observed in the Pt_5P_2 sample despite its polycrystalline nature, the magnetoresistance increases with increasing crystallinity. If we can synthesize a Pt_5P_2 single crystal sample, we can expect to observe a huge magnetoresistance that is much larger than that of existing materials.

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