学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　博士（理学）　　氏名　林　浩章

学 位 論 文 題 名

Magnetism and Hall Effect in Intermetallic Compounds with Noncollinear Spin Textures
（非共線磁気構造を持つ金属間化合物の磁性とホール効果）

The development of functional materials, as represented by sensitive sensors and magnetic memories, is one of the most important topics in a solid-state chemistry. The functionality of such novel devices are mediated by electrons, especially by electrons' spin, and understanding the nature of spins is essential for innovating them. Recently, some of exotic spin textures such as magnetic skyrmions and magnetic multipole had discovered, which has a potential for applying to higher-performance electronic materials. Therefore, these studies have attracted a much attention from both theoretical and experimental perspectives. On the other hand, the mechanisms of these phenomena are still open to discussion, and further basic research is required. In this thesis, to play a role in the development of functional materials, I focus on the magnetic and electric transport properties through unusual spin textures, and discuss their origins.

Chapter 1 shows an overview of this thesis including brief introduction of various spin textures such as skyrmions and magnetic monopole, subsequently provides exotic electrical transport properties and applicational aspects related to their magnetic structure. Then, the several recent researches on novel compounds exhibiting skyrmions and large anomalous Hall conductivity are introduced.

Chapter 2 summarizes the equipments and methods used in synthesis, crystal growth, and physical property measurements, focusing on the basic principles and mechanisms.

Chapter 3 introduces the physical properties on centrosymmetric skyrmion candidates, $GdOs_2Si_2$ ($I4/mmm$), Gd_2NiSi_3 ($P6/mmc$), and $Gd_2Rh_3Al_9$ ($Cmcm$), synthesized by Czochralski method, floating zone method, and high pressure and high temperature method, respectively. Based on the obtained data, a magnetic phase diagram for each sample was established, and their detailed physical properties were summarized as following.

$GdOs_2Si_2$: The magnetic phase diagram is broadly similar to that of $GdRu_2Si_2$, an isostructural compound that features a square skyrmion lattice. I however found a novel phase that does not exist in $GdRu_2Si_2$ just above the double-Q skyrmion phase in the diagram ($B//[001]$). The topological Hall effect observed in the skyrmions phase is almost completely suppressed in this new phase, which strongly suggests that it is a theoretically anticipated double-Q trivial phase. This finding emphasizes the singularity of the topological skyrmion phase in a centrosymmetric lattice system.

Gd_2NiSi_3: The single crystals of Gd_2NiSi_3 have successfully grown by substituting Ni for Pd in a centrosymmetric skyrmion compound, Gd_2PdSi_3. Magnetic measurements reveal that antiferromagnetic transition mediated by Gd^{3+} ions ($S = 7/2$) exhibits magnetic field dependence, and the established phase diagram indicates intriguing similarities to Gd_2PdSi_3. Furthermore, a clear Hall anomaly in Phase II (between $H = 1.2$ and 2.3 T at $T = 2$ K) is consistent with topological Hall effect, suggesting the emergence of skyrmion lattice in Phase II.

$Gd_2Rh_3Al_9$: This intermetallic compound exhibits an orthorhombic structure characterized by a distorted Gd-honeycomb network. Comprehensive investigations of its temperature-dependent behavior in polycrystalline samples reveal sequential antiferromagnetic transitions occurring at $T_1 = 13.6$ K and $T_2 = 4.1$ K. These transitions arise from the antiferromagnetic interaction between the magnetic moments of Gd^{3+} ($S = 7/2$) situated within the distorted honeycomb layer. Despite not observing a skyrmion phase in this compound, the data provide valuable insights into the complex behavior of Gd-based intermetallic compounds and their potential as hosts for novel magnetic phases. Further research, particularly using single crystals, is needed to explore the possibility of forming a skyrmion phase in this compound.

Chapter 4 primely focuses on Mn_3Sb, exhibiting a large anomalous Hall conductivity in the cubic-lattice antiferromagnet with kagome lattice. The measured value is comparable to the theoretical value and more than 50 times larger than the experimental observation for Mn_3Ir (at 300 K). This result suggests that the experimental issues reported so far in cubic antiferromagnet Mn_3X ($X =$ Ir, Pt, Rh) are not essential, and further studies will enhance the possibility of developing new antiferromagnetic spintronic materials characterized by cluster-multipole moments.

Chapter 5 presents the overall conclusions and future prospects based on this thesis.