論文 / 著書情報
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題目(和文)	磁気的フラストレーションを有するクロムスピネル化合物およびパイロクロアチタン酸化物におけるスピン−格子結合に関する熱力学的研究
Title(English)	Thermodynamic Studies of Spin-Lattice Effects in Magnetically Frustrated Chromium Spinels and Titanium Pyrochlores
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論 文 要 旨

THESIS SUMMARY

専攻：Department of	物質電子化学専攻 専攻	申請学位（専攻分野）：Academic Degree Requested	博士 Doctor of （ 理学 ）
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

The concept of *frustration* is attracted much attention as the playground for exploring novel physical properties. A material having a frustration is difficult to achieve a stable ground state, while strong fluctuation emerges at low temperatures. In order to relieve the frustration, the strong fluctuation will be coupled with perturbations, which provides an exotic ground state. In magnetically frustrated systems, the interplay between spin and lattice degrees of freedom, called as spin-lattice coupling, often plays an important role. This doctoral thesis aims to understand the lattice behavior in pyrochlore-based frustrated magnets with the strong spin-lattice coupling. Frustrated compounds are extensively investigated through x-ray and neutron scattering experiments, although the lattice has been assumed to be a passive bystander. Assuming the presence of the strong spin-lattice coupling, the detailed study on the lattice behavior should provide not only insights into lattice properties but also deeper understanding of spin properties. I demonstrate that the thermodynamic study can provide new insights into spin properties in the investigated systems, chromium spinels $A\mathrm{Cr}_2X_4$ (A=Cd, Co, and X=O, S) and titanium pyrochlores $R_2\mathrm{Ti}_2\mathrm{O}_7$ (R=Tb, Dy, and Ho).

In chapter 1, the concept of magnetic frustration, geometrical frustration in the pyrochlore-based magnets, and the spin-lattice coupling is firstly introduced. Then, how the thermodynamic study contributes to the study on the magnetically frustrated pyrochlore-based magnets are presented.

In chapter 2, the experimental background, basic knowledge and conditions are described. This study employed the thermal relaxation method and quasi-adiabatic method for heat capacity measurements, the capacitance method for thermal expansion measurements, and a SQUID magnetometer for magnetization measurements.

In chapter 3, "*spin-glass-like behavior in the ferromagnetic phase* $CdCr_2S_4$" is reported. This compound has been considered to be a typical three-dimensional Heisenberg ferromagnet. The systematic thermodynamic investigation discovered the presence of anomalous behaviors, especially a spin-glass-like behavior below 20 K, in the ferromagnetic phase, which are not expected for the conventional ferromagnet. These observations propose that short-range magnetic clusters arising from the presence of the local lattice distortion grow with decreasing temperature, which would lead to novel physical properties in $\mathrm{CdCr_2S_4}$.

In chapter 4, "*Spin-lattice coupling effect in strongly geometrically frustrated spinel*

$CdCr_2O_4$" is presented. Due to the strong frustration arising from the antiferromagnetic interaction between Cr^{3+} spins, the system undergoes a magnetic transition with a structural transition well below a Curie-Weiss temperature. Thermal expansion measurements found out the peculiar thermal expansion behavior related to its spin behavior, which provides a new insight on the formation mechanism of the spin cluster observed in the paramagnetic phase. In addition, the discussion of the pressure dependence of the transition temperature pointed out the key to clarify the reason of a different transition behavior between $CdCr_2O_4$ and the family compound $ZnCr_2O_4$.

In chapter 5, "*Lattice behavior of the conical spin state in spinel oxide $CoCr_2O_4$*" is described. $CoCr_2O_4$ exhibits a complex sequence of magnetic transitions due to the frustration between Cr-Cr and Co-Cr exchange interactions. Using thermodynamic relations, the stability of each phase, especially for the conical spin state expected to have strong spin-lattice coupling, can be investigated. Furthermore, magnetocaloric measurements in pulsed high magnetic fields observed a first-order transition with large latent heat, and these results determined the magnetic field-temperature phase diagram for the <111> and <001> field directions up to 55 T. These results might indicate that the anisotropic lattice distortion occurs at the lock-in transition.

In chapter 6, "*Lack of the spin-lattice coupling in spin-liquid material $Tb_2Ti_2O_7$*" is discussed. $Tb_2Ti_2O_7$ does not show any conventional long-range order down to 20 mK, which suggests the realization of the spin liquid state. To understand its mechanism, recent studies begin to suggest that the spin liquid state is stabilized by the presence of the spin-lattice coupling. However, the analysis of the heat capacity and thermal expansion coefficient shows that the spin liquid ground state might be established with little relation with a spin-lattice coupling. The enigmatic ground state of $Tb_2Ti_2O_7$ is also argued from the results of heat capacity measurements under high magnetic fields up to 17 T.

In chapter 7, "*Thermal expansion behavior in spin-ice state in $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$*" is studied. The Dy^{3+} and Ho^{3+} ions behave as an ferromagnetic Ising spin with the cubic <111> anisotropy, which results in highly frustrated ground states with spin ice configuration on each tetrahedron. An anomalous thermal expansion behavior was found in the spin ice regime of $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$; interestingly, $Dy_2Ti_2O_7$ shows negative thermal expansion, while $Ho_2Ti_2O_7$ positive thermal expansion. The observed opposite behavior is discussed in terms of the spin ice state. The careful consideration leads to the hypothesis that negative thermal expansion is the common property in the spin ice compound, suggesting the new analogy between spin ice and water ice.

Finally, in chapter 8, summary and concluding remarks are presented.