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博　士　論　文　の　要　旨

専攻名　システム創成科学専攻

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博 士 論 文 題 名 **Study on magnetism in kagome-lattice compound $MgMn_3(OH)_6Cl_2$**

(カゴメ格子物質 $MgMn_3(OH)_6Cl_2$ の磁性に関する研究)

要旨（２，０００字程度にまとめること。）

The phenomenon of magnetism in solids has received a lot of interest since ancient time because of scientific and technological importance. The advancement in the field of magnetism has started from the studies of strongly correlated electron system in which the collective interaction between the electrons are treated by quantum mechanical effect and provide the opportunity to find novel states of matter. The geometrical frustration which is supposed to arise due to lattice geometry of the crystal for which the coupling between the lattice and the spin make it difficult for a system to gain lowest possible energy states has been playing a central role to induce such novel states of matter. The competition between the lattice frustration and the exchange interaction in the frustrated system results unconventional ground states like spin liquid, spin ice, spin nematic, and spin glass, etc. [1-3]. Geometrical frustration is usually found in 3D-pyrochlore lattice, triangular lattice, and kagome lattice geometry. The kagome lattice is suitable to understand the noble states like spin liquid in solids because of having large degenerate ground states in compared to other lattice geometry. The constituents spin in the spin liquid are highly correlated but fluctuating strongly down to low temperature. Spin liquid can be classical and quantum depending on the spin moment. The experimental observation of S = 1/2 quantum spin kagome herbertsmithite $ZnCu_3(OH)_6Cl_2$ has been treated as the first practical spin liquid materials which shows higher frustration [4-5]. A lot of research has already be done on $ZnCu_3(OH)_6Cl_2$, there are many unanswered issues still be present related to the ground state of S = 1/2 kagome antiferromagnets. On the other hand, the Ising-like S = 3/2 kagome antiferromagnets $ZnCo_3(OH)_6Cl_2$ and $MgCo_3(OH)_6Cl_2$ showed short-range correlations with persistent spin fluctuations in the vicinity of the ordered state below T = 2.7 K [6-7]. The S = 2 $MgFe_3(OH)_6Cl_2$ [8] compound behaved like a Heisenberg spin system showing a 120º nearest-neighbour long range order (LRO) with spin vector chirality q = -1. These experimental observations clearly show a tendency from spin liquid to LRO with increased magnetic moment. Almost all theoretical studies predicted an extremely high degeneracy of ground state for classical spin kagome antiferromagnets. But there is a bigger discrepancy between the theoretical prediction and experimental result for classical kagome antiferromagnets as compared to the quantum spin kagome system. Therefore, the ground state of the classical kagome antiferromagnet $MgMn_3(OH)_6Cl_2$ is of

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considerable interest. Motivated by the above discussion, the author would like to synthesize polycrystalline classical kagome antiferromagnets $Mg_xMn_{4-x}(OH)_6Cl_2$ by solvothermal reaction process.

The white coloured polycrystalline $Mg_xMn_{4-x}(OH)_6Cl_2$ compound has been successfully synthesized by selectively substituting the triangular-lattice-plane Mn^{2+} with non-magnetic Mg^{2+}. The compound crystallizes in rhombohedral structure in space group *R*-3*m*, in a similar crystal structure to the much-researched quantum spin liquid candidates herbertsmithite $ZnCu_3(OH)_6Cl_2$ and tondiite $MgCu_3(OH)_6Cl_2$. The temperature dependant magnetic susceptibility reveals antiferromagnetic transition below T_N ~ 8 K. The neutron powder diffraction experiment confirmed long-ranged magnetic order developed below T_N ~ 8 K. The obtained value of critical exponent, β = 0.35, agrees with a 3D Heisenberg spin system with 120° nearest-neighbor spin structure confined in the kagome plane with spin-vector chirality of q = +1 below the transition temperature. Since the transition temperature in the $Mg_xMn_{4-x}(OH)_6Cl_2$ compound lies between 7.6 - 8 K, present work suggests the intrinsic nature of long-range order in classical Heisenberg kagome antiferromagnet and provides a classical reference system to quantum kagome antiferromagnets.

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