Chapter 7

CONCLUSIONS AND FUTURE SCOPE

7.1 Conclusions

Frustrated systems help to discover new states and new properties of matter. $Ho_2Ge_xTi_{2-x}O_7$ series had been synthesized through standard solid-state route. $Ho_2Ti_2O_7$ crystallizes in $Fd\bar{3}m$ space group and $Ho_2Ge_2O_7$ belongs to tetragonal lattice class having $P4_12_12$ space group. The crystal lattice is bipartite consisting of two interpenetrating sublattices. $Ho_2Ti_2O_7$ and $Ho_2Ge_2O_7$ magnetically frustrated system are important due to their exotic spin relaxation phenomenon originating from their ground state degeneracy. The possibility that it is lifted by quantum fluctuations along with the correlations within the ground states and low temperature spin dynamics had been discussed in details. The role of chemical pressure upon the band gap tenability from insulator class of systems ($Ho_2Ge_2O_7$) to high band semiconductor class ($Ho_2Ti_2O_7$) had been established.

The key findings of this thesis work could be summarized as follows:

1. $Ho_2Ge_xTi_{2-x}O_7$ (cubic end) pyrochlore magnetic materials constitute a new class of robust spin ice system. Antiferromagnetic exchange interaction, J_{nn} controls the spin dynamics effectively and more sensitive to the structural distortions. Tunneling dynamics at T_{ice} (~ 2 K) in $Ho_2Ti_2O_7$ is its intrinsic consequence because of prevailing geometrical frustration.

2. In $Ho_2Ge_2O_7$, which crystallize in tetragonal lattice ($P4_12_12$ space group), low-temperature short-range spin correlation have ferromagnetic origin. Ferromagnetic correlations decrease with increase in negative chemical pressure effect in $Ho_2Ge_2O_7$. Single ion spin freezing around 14 K is prevalent in this spin disordered system putting this system under the class of classical spin-ice system.
3. The upper valence band for $Ho_2Ge_xTi_{2-x}O_7$ consisted of O-2p state hybridized with Ho-5p and Ti-3p and Ge-4p states whereas conduction band primarily formed by Ho-5d state hybridized with Ti-3d and Ge-4d states as obtained from Density of States (DOS) calculations. The evolution of total DOS in $Ho_2Ge_xTi_{2-x}O_7$ shows that valence band edge is more sensitive than conduction band to composition. Band gap of 5.24 eV for $Ho_2Ge_2O_7$ drastically drops to 3.92 eV with immediate Ti^{4+} substitution at Ge site in $Ho_2Ge_2O_7$. A high probability of 4f–4f forbidden optical transitions corresponding to Ho states is present in these systems which suitably places these systems in the category of having potential application in optical devices. These results provide chemical pressure as an excellent tool to tailor the band gap in $Ho_2Ge_xTi_{2-x}O_7$.
4. The electronic structure has been calculated from density functional theory using local density approximation. Excellent agreement between the theoretical and experimental values has been obtained in this study.

7.2 Suggestions for future work

There are few open questions that remain to be investigated in $Ho_2Ge_xTi_{2-x}O_7$ as summarized below.

1. A unified magnetic phase diagram for the precise orientation for short-range spin correlation needs to be clarified through a detailed analysis of the magnetic structure using neutron diffraction experiments.
2. In future, it would be interesting to see the crystallographic changes of such systems at relatively much lower temperatures, where spin ices are known to freeze and fall out of equilibrium.
3. However, most of the possible states as predicted by theory remains largely uncharted at an experimental level. Immaculate realization of materials required to investigate interpretative behavior is still lacking. Hence, new systems need to be synthesized to tune the dipolar and exchange interactions for inducing long range ordering which could play a leading role in the future of frustrated magnetism.