Quantum Spin Ice for Pr Pyrochlore Magnets

SHIGEKI ONODA, YOICHI TANAKA, Condensed Matter Theory Laboratory, RIKEN — We theoretically propose a new state comprising a quantum-mechanical analogue of the spin ice for pyrochlore magnets. In classical spin-ice systems like Dy$_2$Ti$_2$O$_7$, the ice rule is mainly driven by the magnetic dipolar interaction, which is proportional to the square of the total angular momentum $J$. Therefore, for Pr$^{3+}$ ions having two $f$ electrons forming the $J = 4$ localized moment, the dipolar interaction becomes an order of magnitude smaller than that for Dy ions. Then, the magnetic superexchange interaction should play a important role. In fact, the form of the exchange interaction is nontrivial because of the highly relativistic nature of $f$ electrons with strong LS coupling and crystal-field effect. Here, we present a microscopic derivation of the effective relativistic spin-orbital Hamiltonian for the pyrochlore magnets Pr$_2$TM$_2$O$_7$ with a transition-metal element $TM$. Then, it is shown that the nearest-neighbor exchange interaction is significantly modified from antiferromagnetic to ferromagnetic by quantum-mechanical processes through excited states split by the crystal field. This bears a quantum-mechanical formation of the ice rule for Pr magnetic moments. Solving the Hamiltonian for a Pr$_4$O tetrahedral cluster, we obtain a further small energy splitting of the low-energy states, leaving doubly degenerate ground states per tetrahedron. Implications for the lattice model and possible relevance to experiments are also discussed.