Spin-orbital frustration in pyrochlores $A_2Mo_2O_7$

Hiroshi SHINAOKA [1], Yukitoshi MOTOME[2], Takashi MIYAKE[3], Shoji ISHIBASHI[3]

[1] *ETH Zürich, Switzerland*

[2] *University of Tokyo, Tokyo, Japan*

[3] *AIST, Tsukuba, Japan*

Mo pyrochlores $A_2Mo_2O_7$ have been attracting much attention due to their fascinating electronic and magnetic properties. In particular, insulating compounds (*A*= Y and Tb, *etc*.) exhibit a spin-glass (SG) transition instead of conventional long-range ordering. The SG behavior has been studied by using isotropic Heisenberg antiferromagnetic (AFM) spin models on a pyrochlore lattice. There, the origin of SG was attributed to the AFM interactions under geometrical frustration. However, recent neutron scattering experiments for *A*=Y [1] revealed diffuse FM magnetic scattering at low *T*. The coupling between orbital and spin moments was also pointed out by first-principles calculations [1]. These indicate a need to reconsider the microscopic picture of the magnetism.

We study electronic and magnetic properties of $Y_2Mo_2O_7$ by the fully relativistic density-functional theory plus on-site repulsion (*U*) method [2]. We show that the system exhibits peculiar competition in energy between different AFM and FM states in an insulating phase. We show that the effective spin interactions are distinct from the simple Heisenberg form and strongly anisotropic in spin space. The keen competition of anisotropic spin interactions gives rise to competing AFM and FM spin correlations at low *T*. Analyzing a three-orbital Hubbard model, we clarify that the magnetic competition is tightly connected with orbital frustration in the $4d^2$ electronic configuration through the relativistic spin-orbit coupling. The results well explain the recent neutron experimental results, and challenge the conventional SG picture based on the geometrical frustration of purely AFM Heisenberg exchange interactions.

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