Ab-initio Green's function methods for modeling magnetic compounds with strong spin orbit coupling

Motivation:

Magnetism is perhaps the oldest collective quantum phenomenon used in practical applications, having been employed in compass needles for over 2000 years. The ancient Chinese, without knowing the underlying quantum properties, used lodestone or magnetite to find the orientation of the Earth's magnetic field. Since then, numerous quantum materials exhibiting phase transitions to states with long-range order, such as ferro-, ferri-, antiferromagnetic, and most recently altermagnetic materials, have been discovered.

Magnetic materials enable a myriad of technological applications. They are essential components of speakers, electric motors in household appliances, inductive generators in wind turbines, and regenerative brakes in electric cars. In modern computer technology, they are used for information-storage devices. However, despite their enormous technological importance for established and emerging technologies, progress in developing new permanent magnets with improved properties, discovering new molecular magnets with desired properties, or searching for new altermagnetic materials has not been straightforward. This is because magnetic compounds are usually synthesized by inorganic chemists either as solids or as single molecule magnets using an empirical approach, which constrains the design of materials with targeted structures and properties.

Scientific Challenges:

The development of modern theoretical ab-initio methods describing magnetic materials could help inorganic chemists optimize their progress. Unfortunately, theoretical modeling of magnetic materials is rife with challenges. One has to accurately describe how electrons correlate in these materials, how temperature affects transition temperatures and magnetic properties, and finally, incorporate relativistic effects such as spin-orbit coupling (SOC), especially since these materials usually contain heavier transition or lanthanide atoms. Methods such as density functional theory (DFT), which are vastly successful in solid-state calculations, face many challenges in delivering predictive results for these materials. Ab-initio quantum chemical methods based on wave function approaches usually become too costly to apply to solids and large molecular magnets.

Description of Proposed Research:

In this proposal, we will develop ab-initio, chemically accurate Green's function methods applicable to both molecular magnets and solid-state regimes. We will implement an extended atomic mean field exact two-component GW formalism to accurately describe relativistic effects such as SOC. Building on this development, we will create a Green's function embedding approach capable of accurately describing strongly correlated electrons in d- and f-orbitals in the presence of SOC. All these advancements will be implemented in an open-source periodic code called GREEN, which will be publicly available.

Finally, we will apply these computational tools in theoretical investigations of the newest altermagnetic materials, spin–orbit coupled molecular quantum magnets realized in inorganic solids, and more traditional antiferromagnetic materials or molecular magnets synthesized by our experimental colleagues.