

Proposed Teaching Activity

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Title of course: *Molecular magnetism and molecular spintronics: a theoretical and computational perspective.*

Duration: 30 hours, partitioned in 3 modules of 10 hours each (8 lectures and 2 tutorial/practical sessions per each module);

Prerequisites: Basic quantum, statistical and chemical physics.

Planned organized times: 1+2 months arrangement is proposed: the first module will be taught during last month of Semester 1 (December 2014); the last two modules during the last two months of Semester 2 (April and May 2015).

Synopsis: The course will introduce the basic theoretical aspects underlying the magnetic and spin-transport properties of single-molecule magnets (SMMs), which are open-shell metal-organic molecules displaying hysteretic spin-dynamics at finite temperatures originating at the single-molecule level, making them promising candidates for the storage and processing of information at the single-molecule level. The models needed to describe the interplay between strong electron correlation, spin-orbit coupling, crystal field effects, spin-phonon coupling and quantum tunneling phenomena in SMMs will be introduced, and used to discuss a set of key experiments probing the magnetism and spin dynamics in these systems. In the final module of this course we will present the basic mechanisms of charge and spin quantum transport in interacting nanodevices, and use them to discuss the possibility of probing and controlling the spin dynamics in SMMs in non-crystalline environments via charge and spin currents.

Learning outcomes: The student will learn how to set up and solve mathematical models based on molecular quantum mechanics to determine the magnetic states of single-molecule magnets, and assess their static and dynamic magnetic properties at finite temperatures. These models will assist the student in simulating experiments in molecular magnetism and molecular spintronics, such as DC- and AC-magnetometry, EPR and paramagnetic NMR spectroscopies, inelastic neutron scattering and magneto-transport experiments, both within a molecular break-junction set up, and in hybrid devices with SMMs grafted on the surface of a current-carrying nanodevice.

Course plan:

Module I (first semester: December 2014)

Lectures (8 sessions):

- A bird's-eye view of molecular magnetism: experimental evidence, heuristic theoretical descriptions, and common knowledge.
- Brief review of free-ion magnetism: angular momentum theory; atomic terms; spin-orbit coupling; atomic multiplets; Hund's rule magnetism; Zeeman effect.
- From magnetic atoms to magnetic molecules: introduction to crystal field theory.
- Magnetic anisotropy in molecules; zero-field splitting effective spin Hamiltonian; classical electrostatic origin of the uniaxial magnetic anisotropy in lanthanide single-ion complexes: heuristic evidence and open questions.
- Equilibrium observables in molecular magnetism: free-energy perturbation theory; molecular magnetization; Van Vleck theory of magnetic susceptibility; EPR g-tensor, and paramagnetic-NMR chemical shifts.
- Exchange interactions in magnetic molecules, and Heisenberg exchange effective spin Hamiltonian. Effects of magnetic anisotropy and orbital degeneracy on magnetic exchange.
- Put it all together: fundamental microscopic ingredients to achieve single-molecule magnets (SMMs) and single-ion magnets (SIMs). Ab initio quantum chemistry approach to the study of SMM's and SIM's.
- Exotic magnetic states in molecules: interplay between exchange interactions, magnetic anisotropy and spin frustration, and emergence of toroidal magnetic moments (*a.k.a.* magnetic anapole moments) and chiral spin states.
- Challenges from the theoretical/computational perspective.

Tutorials Module I (2 sessions): we will work out the details of some of the models introduced at lectures, and use them to discuss some example of experimental interest in molecular magnetism.

Module II (second semester: April 2015)

Lectures (8 sessions):

- Time-dependence in quantum theory at finite temperatures: the T-matrix approach, and the density matrix formalism.
- Equations of motion for the reduced density matrix of open quantum systems.
- Spin-phonon coupling and spin-lattice relaxation.
- Dynamic fingerprints of SMMs: mechanisms of slow magnetic relaxation in time-dependent magnetic fields and blocking temperatures. Quantum tunneling of the magnetization.
- Linear-response: ac-susceptibility; origin and signature of Arrhenius law and the single-barrier activated slow relaxation; signature of quantum tunneling;
- Beyond linear response: generalized master equation description of magnetic hysteresis in SMMs.
- Challenges from the theoretical/computational perspective.

Tutorials Module II (2 sessions): We will work out the details of some of the models introduced at lectures via a series of theoretical and numerical exercises, and use the results to discuss a few examples of experimental interest in molecular magnetism.

Module III (May 2015)

Lectures (8 sessions):

- Basic coherent quantum transport: the scattering matrix approach; transfer matrix formalism; the Landauer formula.
- Time-dependence in quantum theory at finite temperatures: the equilibrium Green's function approach.
- Equations of motion for equilibrium Green's function. Example: the Anderson model for magnetic impurities.
- Non-equilibrium quantum transport in interacting mesoscopic systems: non-coherent (Coulomb blockade) and coherent (co-tunneling & Kondo) regimes.
- SMMs devices in the sequential tunneling regime: interplay between spin currents, magnetic anisotropy and mixed-valence redox states.
- Time-dependent quantum transport: probing molecular magnetism in non-crystalline environments via spintronics.
- Challenges from the theoretical/computational perspective.

Tutorials Module III (2 sessions): We will work out the details of some of the models introduced at lectures via a series of theoretical and numerical exercises, and use the results to discuss a few examples of experimental interest in molecular magnetism and SMM-based spintronics.