Ab initio theory for ultrafast spin dynamics

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This tutorial will start with a short historical overview of the group's achievements with regard to the different time scales for the dynamics on solids. The effects studied include (from the fastest to the slowest) bleaching [1,2], dephasing [1,3], population dynamics [4,5], electron-phonon coupling [6], and spin-lattice relaxation [7].

After a short motivation an overview of the theoretical consideration leading us to choose molecular systems with two and three active magnetic centers will be given. In particular we will explain the importance of the number of active magnetic centers for coherent spin manipulation and functionalization of magnetic nanostructures [8]. In order to realize optical spin manipulation we apply subpicosecond laser pulses on carefully chosen materials. The correlated electronic and magnetic properties of the chosen molecular magnets are treated from first principles within the symmetry-adapted cluster configuration interaction method. Spin-orbit coupling and a static magnetic field are included perturbatively. The coherent spin control is performed with two optical transitions forming a Lambda process.

In the next part some of the structure families studied will be presented: linear chains, ligand-stabilized magnetic complexes, small on surfaces, and clusters in the gas phase.

The first family consists of linear chains with two magnetic centers at their end (Co, Ni, Fe) linked with nonmagnetic metallic Na atoms [9,10]. Our main findings are: (i) that the spin density is mainly localized on a single magnetic center for most of the low-energy electronic states, (ii) a local spin-flip is almost always possible within less than a picosecond, and (iii) that the Fe, Co and Ni centers exhibit different magnetic switching behavior: Fe switches with one Rabi oscillation, Co in a cascade-like manner, while Ni shows a much more complicated behavior [9].

Next an already synthesized and experimentally characterized two-magnetic-center ligand-stabilized complex is presented, which carries the minimum ingredients for all-optical ultrafast spin-manipulation, namely two Ni atoms with different local symmetries (one Ni being in an octahedral field and the other in a square planar ligand configuration), and hence different multiplicities, and localized spin density [11]. For this molecule a spin-switch scenario has successfully been calculated.

Third we move to a family of magnetic nanostructures which consist of three magnetic centers and are deposited on hypothetical surfaces. For some of these structure we have realized both local spin-switching and spin-transfer scenarios, and, further than that,

successfully combined those scenarios to produce functional logic elements (AND, OR, and XOR gates) [12].

Fourth, the last family presented consists of small metallic clusters in gas phase. On these we have also succeeded in realizing spin-switching and spin-transferring scenarios. Additionally, by the use of attached chromophores we show that it is possible to indirectly assess the magnetic state of the systems with conventional infrared spectroscopy [10].

Using the fourth family we investigate the role of the metallicity of the bridging atoms with respect to the degree of spin-localization and spin-transferability [13]. We thus derive five rules-of-thumb: (i) spin switching is easier in the order Fe (easiest) > Co (moderate) > Ni (hardest), (ii) O as a nonmetallic and Mg as metallic prototypic bridging atoms can reduce and relocate maximum of spin density, (iii) spin flip is easier on linear structures, (iv) both Mg and O as bridging atoms increase the efficiency of spin transfer, and (v) we explicitly show the possibility of coherent control (different laser pulses lead to different results) which paves the way for more structures that can be logically functionalized.

Last but not least we will give a short outlook of new structures and control mechanisms that can be of outmost interest within the frame of molecular magnetism.

Literature

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