Phono-magnetism

Coupled spin nuclear dynamics and its impact on Magneto Optical Kerr Effect, Magnetic Circular Dichroism and Photo Emission Spectra

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In Short

- The questions that haunt the field of femtomagnetism are (a) What is the impact of the spin (and pseudo spin)-dynamics on nuclear dynamics, and vice versa, in laser induced dynamics of extended systems? (b) Can we tune the laser pulses to excite specific nuclear dynamics in order to better manipulate (pseudo) spins at ultra-fast time scales? and (c) What is the signature of nuclear dynamics on response functions like transient magneto-optical Kerr effect (MOKE), transient magneto circular dichroism (MCD)?
- This project aims to answer these questions by means of coupled charge and spin dynamics with nuclear dynamics. The dynamics of charge and spin will be treated with ab-initio method of time-dependent density functional theory and nuclei will be treated classically.
- This formalism will be applied to 3D ferro- and antiferro magnetic sytems such as FePt, FeMn etc. We will also study the impact of laser parameters on valley- and spin- selective excitations in 2D materials (such as WSe_2 and MoS_2) in the presence of nuclear dynamics.

Ground-state density functional theory (DFT) has, over the last four decades, revolutionized materials science by offering parameter-free predictive insight into material properties. The time-dependent extension of this theory, time-dependent density functional theory (TDDFT), now stands on the cusp of a similar revolution for ultrafast laser-induced spin dynamics. TDDFT has already demonstrated predictive power for early time (less than 100 femtoseconds) spin and charge dynamics, both answering old questions, for example the physical origin of spin dynamics in elemental magnets2, as well as uncovering unexpected new phenomena, such as the optically induced spin transfer (OISTR) effect 15.

In the field of femto-magnetism usually an indirect comparison between theory and experiments is made; the normalized experimental magnetic circular dichroism (MCD) signal is compared to the theoretically calculated magnetization density. While theoretical calculations of ultrafast spin dynamics

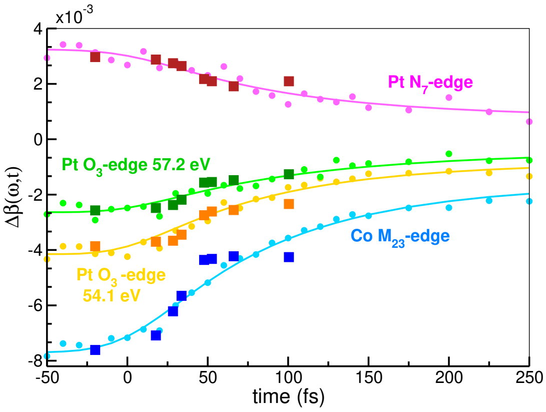

Figure 1: *Various edges in transient-MCD for CoPt alloy. Theoretical data is shown as squares and experimental data as circles. Lines are just guides to the eye. This data was obtained by combining real time TDDFT and linear response TDDFT. An excellent agreement between theory and experiments indicates that approximations used to calculate the response function are valid. Furthermore, early femtosecond (<100fs) dynamics is dominated by OISTR and hence theoretical simulations can in future be used to design a faster method of spin manipulation by pulse and material design3.*

naturally yield spin and orbital moments as a function of time. To bring theory3 a step closer to experiments we developed within TDDFT a method to calculate time-dependent MCD which was then compared to experimental data4 showing excellent agreement between theoretical and experimental MCD spectra (see Fig. 1). This indicates that the TDDFT approach can thus now be used to disentangle the early time physics of spin dynamics in complex magnetic systems. The capability to both study laser-induced spin dynamics from first principles as well as to calculate precisely the same transient observables as experiments puts ab-initio theory in a unique position to be able to be predictive and at the same time be able to help provide insight into the experimental data.

While treatment of the spin and charge dynamics for extended systems via TDDFT is now becoming a well established theoretical tool, the coupling of the nuclear and spin degrees of freedom remains challenging within the current state-of-the-art TDDFT for extended solids. Such coupling of nuclear and spin dynamics is, however, crucial for extending the time window of TDDFT beyond the very early times of < 100fs. With such a coupled dynamics we will address following open and fundamental questions: (i) What is the effect of nuclear dynamics on spin dynamics in early times (< 60fs)?

(ii) What laser parameters play the most important role in valley- and spin- selective excitations in 2D materials?
(iii) What are the signatures of nuclear dynamics on response function like transient MOKE and MCD?

It order to couple nuclear dynamics to spin and charge dynamics we plan to calculate forces that act on nuclei due to laser-induced changes in the charge density. Having these forces in hand and treating nuclei as classical particles we will then use Ehrenfest dynamics for moving the nuclei. These new nuclear positions can then be used to time-propagate electronic charge and spin degrees of freedom. The upshot of this is that we will be able to study the effect of laser induced spin- and charge-dynamics on nuclear degrees of freedom. More importantly, we will also be able to study the effect of non-adiabatic nuclear-charge coupling on the spin-dynamics. Finally, with these tools in hand we will aim to control spins not just by direct light-charge coupling mechanism, but also by indirect light-nuclei-charge-spin coupling mechanism.

For the study of phonons we require supercells and simulation times of 20-200fs to include at least one oscillation of a high symmetry mode. Then for this mode, we will run several different pulses, with and without the phonon dynamics, to investigate the influence they have. Given large number of atoms in a unit cell and large number of time-step required a large computer cluster is needed.

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More Information

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