

Theoretical simulation of pump-probe experiments

Theoretical simulation of the probe in pump-probe experiments

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In Short

- One of the most significant impediments to understanding ultrafast spin-manipulation is that the most common experimental tools of choice – Magneto-optical Kerr effect (MOKE), X-ray magnetic circular dichroism (XMCD), and photoemission spectra (PES) - remain beyond the reach of ab-initio simulation.
- This project aims to rectify this by developing an ab-initio theory of these experimental techniques, i.e., a theory of time-resolved MOKE, XMCD and PES and implementing this theory in a state-of-the-art time-dependent density functional theory (TD-DFT) code.
- With the above tools the project aims to (a) elucidate the underlying physical mechanisms behind the ultrafast dynamics of spin and charge and (b) study and design new materials such as ferromagnetic/anti-ferromagnetic (FM/AFM), anti-ferromagnetic/non-magnetic (AFM/NM) and FM/NM interfaces with a view to optimally control spin by light.

Ultra-fast light-induced demagnetization was demonstrated in 1996, where demagnetization times (in Ni) faster than a few picoseconds were achieved using intense electromagnetic pulses. Recently, these spin-manipulation times have been reduced to a few femtoseconds, owing to great advances made in the production of short-time laser pulses. However, we are still far from achieving sufficiently controlled manipulation of spins required for production of useful devices. One of the main reasons behind this is the lack of full understanding of the phenomena leading to demagnetization. There have been a number of attempts at explaining this optically induced spin-dynamics, however, all these attempts to understand the physics of spin modulations by light have one thing in common - they are all based on model Hamiltonians. A critical problem then arises: models with totally different underlying physical assumptions are all able to explain the same experimental data. How then can one determine the fundamental physics of demagnetization?

We will address following two open and fundamental questions:

- (i) What is the physical mechanism behind ultra-fast

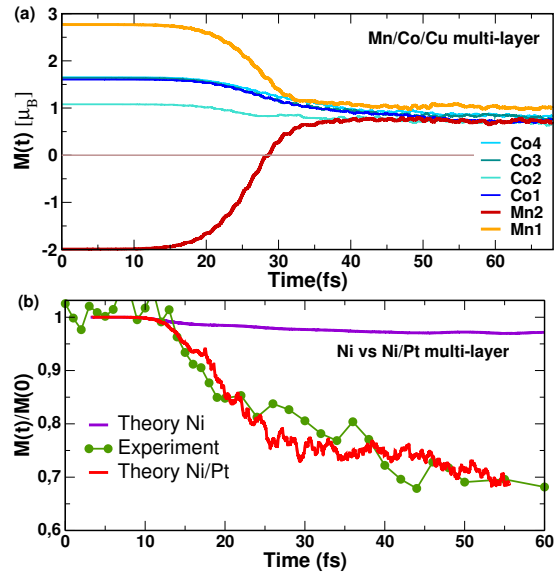


Figure 1: (a) Laser induced ultra-fast change of magnetic order: time evolution of the layer resolved local magnetic moment in a Co/Mn multilayer, which at $t = 0$ fs (i.e., in the ground state) is an anti-ferromagnet. At ~ 29 fs the interface Mn layer – that is coupled anti-ferromagnetically to all other layers at $t = 0$ – demagnetizes, then re-magnetizes but with the spin orientation reversed. The magnetic order thus switches from uncompensated anti-ferromagnetic to transient ferromagnetic. (b) Normalized magnetic moment of Ni/Pt multi-layers and bulk Ni as a function of time. In first 20fs OISTR induces ultrafast loss in the moment in multilayers while this effect is missing in bulk Ni??.

light-induced spin dynamics in various materials, is there is a universal mechanism for ultra-fast light-induced demagnetization, and (ii) What is the difference between various kinds of magnetically coupled systems when it comes to the dynamics of spins – for example FM/AFM, NM/FM and AFM/NM interfaces.

What one requires in such a situation is a fully ab-initio theoretical description, free from any model assumptions. Time-dependent density functional theory (TD-DFT), which extends density functional theory (DFT) into the time domain, is a formally exact method for describing real-time dynamics of charge and spins under the influence of an external field such as an applied laser pulse. In our recent work using TD-DFT we predicted an entirely new effect in ultrafast demagnetization of multicomponent magnets – the type of coupling between the constituent atoms of a magnetic solid, usually ferromagnetic (FM, i.e. with parallel magnetic moments on neighboring atoms) or anti-ferromagnetic (AFM, anti-parallel orientation), is a fundamental property

of any magnetic material. This coupling is governed by the exchange interaction between the electrons on the neighboring atoms. For a typical magnetic material, the timescale associated with this interaction is of the order of a few 100s of femtoseconds. We predicted that it is possible to control magnetization via light pulses even at sub-exchange time scales of the order of few tens of femtoseconds. This includes changing the magnetic order from AFM to FM in materials?? that consist of different types of atoms, forming so-called sub-lattices in the solid. The electrical field of the light pulse was found to induce an electron and associated spin current flowing between the sub-lattices, leading to a transient change of the magnetic order (see Fig.??(a)). Furthermore, it was seen in theory that this optical inter-site spin transfer (OISTR) dominates?? the early time spin-dynamics in all multi-sub-lattice magnetic materials??, followed by spin-orbit coupling mediated processes at slightly later times. This prediction has recently been confirmed experimentally for the Ni/Pt sublattice where atto second MCD experiments demonstrated?? that OISTR is the dominating mechanism in first 10fs. Due to the lack of second sublattice in pure Ni slab this effect was found to be missing (see Fig. ??(b) for details).

It order to take theoretical description of light-matter interaction another step closer to experiments we will extend the present simulations to pump-probe technique as done in experiments. We have developed the Elk code (elk.sourceforge.net) to be able to calculate observables like magnetic dichroism and kerr rotation as a function of time. The quantity that is accessible via TDDFT calculations is the current density, $J(\mathbf{r}, t')$, where the t' refers to the pump pulse. For calculating this current no assumption is made and it encapsulates all non-linear effects coming from the presence of an intense pump-pulse. In presence of a probe pulses this current gets an extra time index i.e. $J(\mathbf{r}, t', t)$. This current can then be integrated over the spatial co-ordinate to get purely time-dependent current

$$j(t', t) = \int_v d^3r J(\mathbf{r}, t', t),$$

where v is volume of the unit-cell. Partial Fourier transform of this current density is $j(\omega, t)$. Under the assumption that the probe pulse is weak and the response of the system to this probe pulse is linear one can write

$$j(\omega, t) = \sigma(\omega, t)E(\omega),$$

where σ is the conductivity tensor and E the electric field vector of the probe-pulse. Such an assumption is perfectly justified because in experiments probe pulses are very weak, usually upto two orders in

magnitude smaller than the pump pulse. Given σ one can then easily calculate the dielectric tensor,

$$\sigma(\omega, t) = \frac{\omega}{4i\pi}(\epsilon(\omega, t) - 1),$$

with ϵ in hand one can calculate both MOKE and MCD. For example the Kerr angle and ellipticity can be extracted by the relation

$$\theta(\omega, t), i\gamma(\omega, t) = \frac{-\sigma_{xy}(\omega, t)}{\sigma_{xx} + \sqrt{(1 + i\frac{4\pi}{\omega}\sigma_{xx})}}$$

Similarly, MCD can be calculated from the imaginary part of ϵ . However, for this two calculation are required– one with left and other with right circularly polarized pump pulse. The difference between the two will then give time resolved MCD

With these tools in hand we will aim towards material and pulse design. One of the most interesting design of pulse example would be: given a AFM system (with small or 0 moment) when excited with a laser becomes FM coupled with large moment. The challenging task would be to design a pulse to make this processes reversible and we will apply optimal control theory in conjunction with TDDFT to do this. Furthermore we would like to explore interfaces FM/AFM or FM/vacuum and AFM/vacuum. One of the example systems of this category is the spin transfer torque where the flow of spin current from one part of the multi-layer to another causes the transfer of spin torque. The details of the spin-current flowing through the junction in these materials is hence of crucial importance. Based on the detail information that we will obtain by our study we will design new multi-layered structures for enhancement of such currents and laser pulses to induce currents.

Given large number of atoms in a unit cell for multi-layers and large number of time-step required for comparison with experiments a large computer cluster is needed.

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<http://www.trr227.de/>

More Information

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