Femto- phono- magnetism

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From the outset of research into femtomagnetism, the field in which spins are manipulated by light on femtosecond or faster time scales, several questions have arisen and remain highly debated: How does the light interact with spin moments? How is the angular momentum conserved between the nuclei, spin, and angular momentum during this interaction? What causes the ultrafast optical switching of magnetic structures? What is the ultimate time limit on the speed of spin manipulation? What is the impact of nuclear dynamics on the light-spin interaction?

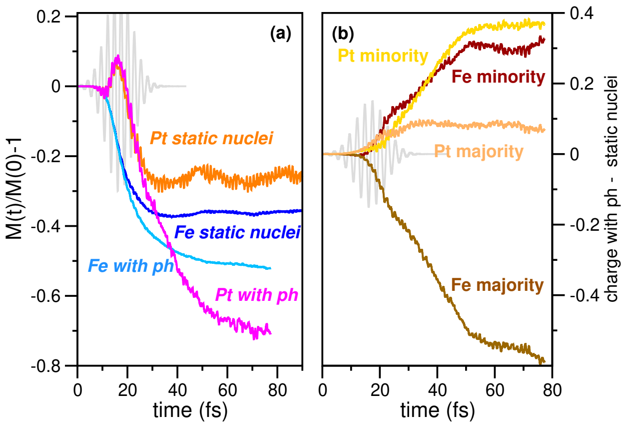

Fig.1 left: (a) Normalized atom-resolved spin moment as a function of time (in fs) in laser pumped (vector potential shown in grey) FePt. The moment is calculated in presence of phonon modes as well as in absence of any nuclear motion. (b) Difference between the excited charge with and without nuclear motion.

In my talk I will advocate a parameter free *ab-initio* approach to treating ultrafast light-matter interactions, and discuss how this approach has led both to new answers to these old questions but also to the uncovering of novel and hitherto unsuspected early time spin dynamics phenomena [1,2]. In particular I will show that selective excitation of optical phonon modes exert a strong influence on femtosecond demagnetisation, generating an additional loss of moment within 40 femtoseconds [3] in laser pumped materials (see Fig. 1). Underpinning this enhanced loss of moment we deduce a new physical mechanism of inter-site charge transfer, dominant in the minority channel. In the second part of the talk I address the question of valley control in 2d transition metal dichalcogenides (TMDC), with current understanding that it couples exclusively via circularly polarized light. In our work we show that on femtosecond time scales valley coupling is a much more general effect. We find that two time separated linearly polarized pulses allow almost complete control over valley excitation, with the pulse time difference and polarization vectors emerging as key parameters for valley control. Finally, I discuss how hybrid pulses combining gap tuned circularly polarized light and linearly polarized THz light I show leads to complete control over valley current states in TMDCs.

References

[1] Dewhust et al., *Nano Lett.* **18**, 1842 (2018).
[2] Siegrist et al. *Nature* 571, **240** (2019)
[3] Sharma et al. *Sci. Advs.* 8, **eabq2021** (2022)
[4] Sharma et al. Optica 9 (8), 947-952 (2022)