Strong-Field-Induced Attosecond Magnetism

Ofer Neufeld, Nicolas Tancogne-Dejean, Umberto De Giovannini, Hannes Hübener, and Angel Rubio

Max Planck Institute for the Structure and Dynamics of Matter and Center for Free-Electron Laser Science, Hamburg, Germany, 22761. oneufeld@schmidtsciencefellows.org

Magnetism is one of the most fundamental physical phenomena in nature. In the past decades, immense efforts were devoted to understanding and manipulating magnetism on ultrafast timescales[1]. However, in all works to date the fastest magnetic response was on the order of few femtoseconds[2,3], and it is not yet clear what the fundamental speed-limit is. Moreover, a magnetic turn-on has not been observed or predicted using linearly-polarized light.

Here we theoretically explore light-driven magnetism in materials that are non-magnetic in their ground state with time-dependent density functional theory (TDDFT)[4]. First, we show that with circularly-polarized driving in the strong-field regime, the system develops an onset of magnetization within a few femtoseconds (see Fig. 1(b)). We find this effect originates from a sub-cycle timescale cascaded transfer process of light's spin angular momentum (SAM) to electronic orbital angular momentum (OAM), which is then transferred to spin angular momentum (through spin-orbit coupling (SOC)) (see Fig. 1(a)). This understanding leads us to propose a new paradigm for inducing attosecond magnetism: By irradiating optically-anisotropic heavy materials with intense linearly-polarized light, purely transient magnetism arises (the slower femtosecond response is suppressed). The induced magnetization then follows the sub-cvcle structure of the electronic orbital angular momentum transfer, which is extremely nonlinear and comprises of high-order harmonics of the laser. Consequently, we predict the fastest magnetic responses to date of ~500 attoseconds, and propose an X-ray circular-dichroism (CD) transient absorption set-up that is capable of measuring such fast phenomenon (Fig. 1(c)). Our work presents a direct path to control the speed of magnetization dynamics down to their potential intrinsic limit - the attosecond motion of electrons in their natural timescale.



Figure 1: (a) Scheme and physical mechanism illustration. (b) light-induced magnetization in circular driving conditions showing femtosecond turn-on of magnetization. (c) Bottom inset is the same as (b), but for linear driving conditions. The 2D map presents simulated pump-probe CD transient absorption, showing that the magnetic response can be observed, including typical ~500 attoseconds oscillations.

References

- [1] Kirilyuk, A., Kimel, A. V, Rasing, T., Rev. Mod. Phys. 82, 2731 (2010).
- [2] Siegrist, F. et al., *Nature* **571**, 240 (2019).
- [3] Okyay, M. S. et al., Phys. Rev. B 102, 104304 (2020).
- [4] Neufeld, O. et al., npj Comp. Mat. 9, 39 (2023).