Direct observation of coherent spin injection across metallic layers at the few-femtosecond timescale

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Femtosecond laser pulses are able to act on the magnetic moments of solids, as amply demonstrated by magneto-optical experiments.^{1,2} However, the vast majority of control schemes make use of incoherent interactions (scattering, diffusion) between electrons, spins and phonons. On the contrary, the observation of direct and coherent interactions between light and spins has been scarce, despite first hints discovered more than ten years ago.³

Here we examine the ultrafast spin response of Co/Pt multilayers when driven by 4 fs laser pulses. The material response is captured by magnetic circular dichroism (MCD) using broadband attosecond pulses. This recently developed approach⁴ is extended here by measuring MCD across both the Pt $O_{2,3}$ and the Co $M_{2,3}$ edges. The method thus combines outstanding time resolution and element-specificity, providing a layer-resolved probe of electronic and spin dynamics.

The experiment shows simultaneous electronic excitation in Pt and Co layers and carrier thermalization times of 8-10 fs. Most intriguingly, it also uncovers a direct spin response to the laser pump pulse: the Pt (resp. Co) magnetic moment transiently increases (resp. decreases) when the driving laser pulse is present (see Figure). After that, both magnetic moments begin to decay with two distinct timescales, ascribed to coherent and incoherent demagnetizing processes. Simulations in the framework of fully noncollinear time-dependent density functional theory (TD-DFT)⁵ reproduce this behavior. Our work constitutes an unambiguous observation of laser-driven injection of majority spins from Co to Pt, offering a way towards realizing spintronics functionalities in an all-optical and remarkably fast way.

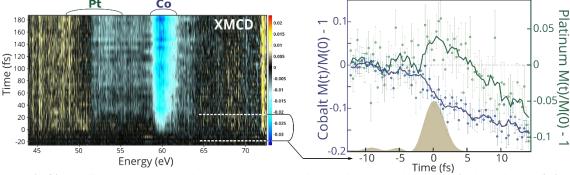


Figure 1: (Left) Broadband magnetic circular dichroism measured across both Pt and Co edges (indicated on top). (Right) Measured dynamics of the Co and Pt magneto-optical index at very early time delays. The brown shaded area indicates the measured pump pulse intensity profile.

- 1. Beaurepaire, E. et al. Phys. Rev. Lett. **76**, 4250–4253 (1996).
- 2. Stanciu, C. D. et al. Phys. Rev. Lett. 99, 047601 (2007).
- 3. Bigot, J.-Y. et al. Nat. Phys. 5, 515-520 (2009).
- 4. Siegrist, F. et al. Nature **571**, 240–244 (2019).
- 5. Dewhurst, J. K. et al. Nano Lett. 18, 1842–1848 (2018).