

# State-resolved ultrafast charge and spin dynamics in [Co/Pd] multilayers

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## ABSTRACT

We use transient absorption spectroscopy with circularly polarized x rays to detect laser-excited hole states below the Fermi level and compare their dynamics with that of unoccupied states above the Fermi level in ferromagnetic [Co/Pd] multilayers. While below the Fermi level, an instantaneous and significantly stronger demagnetization is observed, above the Fermi level, the demagnetization is delayed by  $35 \pm 10$  fs. This provides a direct visualization of how ultrafast demagnetization proceeds via initial spin-flip scattering of laser-excited holes to the subsequent formation of spin waves.

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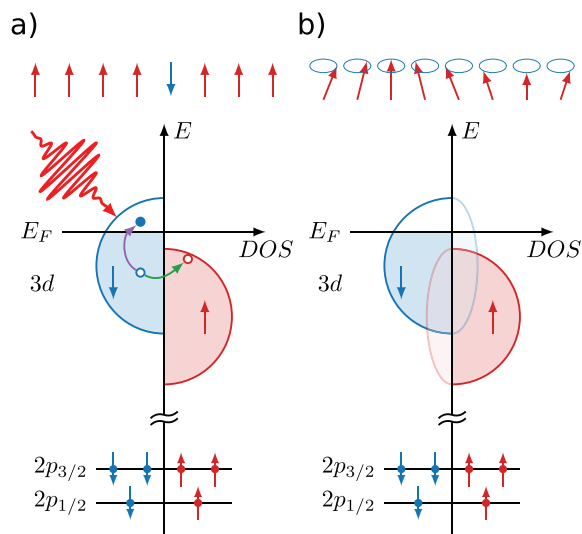
Femtosecond optical excitation of magnetic materials and heterostructures leads to strongly non-equilibrium conditions displaying many novel phenomena that are absent in equilibrium physics (for reviews see Refs. 1 and 2 and references therein). Discoveries, such as all-optical magnetization reversal,<sup>3–6</sup> superdiffusive spin transport,<sup>7–14</sup> and optically induced spin transfer effect,<sup>15,16</sup> not only challenge our

fundamental understanding but also provide future perspectives for information storage and processing. It is generally accepted that laser excitation initially leads to a highly non-equilibrium electron system while conserving the total electron spin polarization.<sup>17–19</sup> Subsequently, electronic thermalization and magnetization dynamics set in that distribute the deposited laser energy over the system's

electron, spin, and orbital degrees of freedom and ultimately to the lattice.<sup>17–22</sup> While such dynamics can be described by a phenomenological three temperature model, the underlying physics at play remains hidden behind *ad hoc* coupling constants.<sup>23</sup>

For understanding the angular momentum flow during ultrafast demagnetization, it is important to disentangle the influence of magnetic interactions, such as exchange and spin–orbit coupling in the non-equilibrium dynamics. While the exchange interaction leads to processes that conserve spin, such as angular momentum transfer between magnetic sub-systems<sup>24</sup> and the excitation of spin waves,<sup>25–28</sup> spin–orbit coupling can cause electronic spin-flips where the corresponding change in angular momentum is transferred to phonons.<sup>29</sup> Such Elliott–Yafet type spin-flip scattering on phonons has been investigated and is considered a key ingredient of ultrafast demagnetization.<sup>20,23,30,31</sup> Flipping electron spins also lead to the excitation of spin waves<sup>32</sup> via exchange scattering. However, the direct observation of spin waves in momentum space and in the time-domain remains challenging.<sup>33</sup> Several studies reported ultrafast excitation of spin waves for instance via a delayed onset of demagnetization.<sup>34–36</sup> This situation is visualized in Fig. 1(a) where the initial spin-conserving laser excitation and subsequent spin-flip processes are depicted. The decay of a flipped electron spin [top of Fig. 1(a)] into spin waves [top of Fig. 1(b)] is thought to take a characteristic time of several 10 fs.<sup>34–36</sup>

Here, we use time-resolved x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) to follow these processes as they evolve in real time in [Co/Pd] multilayers. We show that  $2p \rightarrow 3d$  core-valence transitions can directly probe the spin-polarization of laser-induced holes below the Fermi level. These states display an instantaneous response to fs laser excitation. Surprisingly,



**FIG. 1.** (a) Schematic of the experiment where the unoccupied  $3d$  spin-resolved density of states (DOS) are probed by  $2p$  core-level absorption spectroscopy. Upon excitation by a femtosecond laser pulse, electrons are promoted from below to above the Fermi level,  $E_F$ , in a spin-conserving process (purple arrow). In a strong ferromagnet, such as [Co/Pd], spin relaxation can only occur below  $E_F$  by a hole spin-flip (green arrow). (b) After the localized hole spin-flip excitation, spin waves are generated and correspondingly the spin-resolved DOS are partially mirrored.

$2p \rightarrow 3d$  transitions into unoccupied states above the Fermi level show a demagnetization dynamics that is delayed by  $35 \pm 10$  fs. In addition, laser-induced hole states below the Fermi level display a much stronger demagnetization. These observations are consistent with the notion that spin–orbit scattering in strong ferromagnets is the driving force for ultrafast demagnetization.

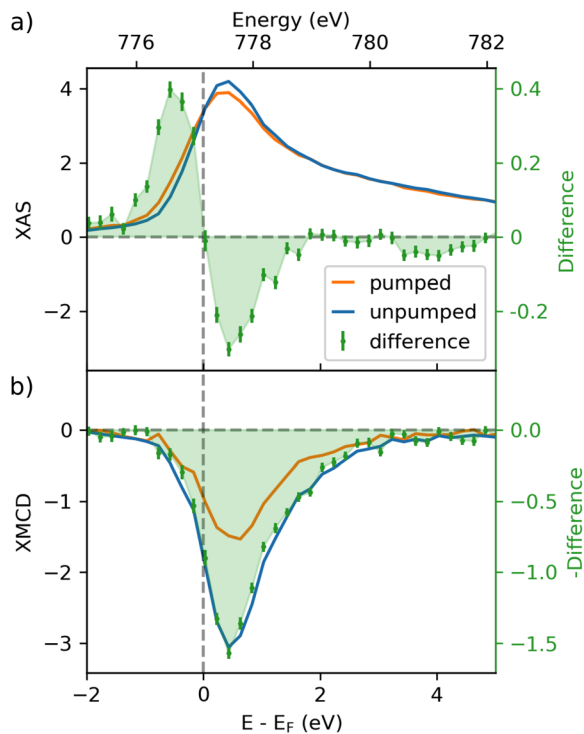
Experiments were performed at the SXR instrument of the Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory. The experimental setup is described in detail in Ref. 37. X-ray absorption spectra were measured in transmission. The incident x-ray intensity was measured via the x-ray fluorescence from a  $\text{Si}_3\text{N}_4$  membrane placed in the beam before the sample and detected with a microchannel plate (MCP). The transmitted x-ray intensity behind the sample was recorded by a fast charge coupled device (CCD) detector. XAS spectra over the  $L_3$  absorption edge corresponding to  $2p_{3/2} \rightarrow 3d$  transitions were acquired by varying the x-ray energy via the LCLS electron beam energy. A 250 meV x-ray bandwidth was selected by the beamline monochromator using a 100 lines per mm grating resulting in an effective resolving power of 3000 at 780 eV.<sup>38</sup> Circularly polarized x-ray pulses were produced using the “delta” afterburner undulator,<sup>39</sup> enabling the measurement of XAS and XMCD spectra by alternating the magnetic field saturating the sample along the beam direction and computing sum and difference for XAS and XMCD, respectively. Time-resolved XAS and XMCD data were acquired by scanning the time delay between the 50 fs Full Width at Half Maximum (FWHM) x-ray probe pulse and the 60 fs FWHM pump laser at a central wavelength of 798 nm. The data were corrected for timing jitter between the pump and probe pulses by measuring the arrival time of the electron pulses via the so-called phase cavity.<sup>40</sup> Slower timing drifts on the few-minutes-scale were corrected using a cross correlation-based time delay estimation method as detailed in the [supplementary material](#). The pump laser was focused on the sample to a spot size of  $190 \times 150 \mu\text{m}^2$  FWHM giving a fluence of  $\mathcal{F} = 35 \text{ mJ/cm}^2$ . The x-ray spot size was  $50 \times 50 \mu\text{m}^2$  FWHM and the x-ray fluence below  $5 \text{ mJ/cm}^2$ .

A  $[\text{Co}(6 \text{ \AA})/\text{Pd}(6 \text{ \AA})]_{38}$  multilayer sample capped with a  $\text{Pd}(20 \text{ \AA})$  layer and grown onto a 100 nm  $\text{Si}_3\text{N}_4$  membrane with a  $\text{Ta}(10 \text{ \AA})/\text{Pd}(30 \text{ \AA})$  buffer layer was used in the measurements described below. The sample was grown by DC magnetron sputtering with fabrication details given in the [supplementary material](#). Prior to the LCLS experiments, the sample was characterized at beamline 4.0.2 of the Advanced Light Source (ALS) using XAS and XMCD measurements, where sum rule analysis confirmed the magnetic properties of the multilayer with previously published work as discussed in the [supplementary material](#).

Conceptually, the experiment is depicted in the schematic shown in Fig. 1. In an itinerant strong ferromagnet, such as Co in [Co/Pd] multilayers, the density of states (DOS) can be separated into completely occupied majority (spin “up”) and partially occupied minority spin (spin “down”) channels, which are shifted in energy by the exchange splitting. At the  $L_3$  absorption edge, valence hole states are probed via  $2p_{3/2} \rightarrow 3d$  core-valence transitions. In the ground state, all electronic states up to the Fermi level  $E_F$  are occupied. As the pump laser pulse excites the  $3d$  electronic system by promoting electrons from below to above the Fermi level, transient XAS can detect the additional hole states below  $E_F$ . XAS transitions into states above  $E_F$ , however, are reduced by the laser-excited transient electron

population in these states. Exactly at  $E_F$ , no XAS changes should be observed. Laser-excited holes below  $E_F$  are thought to lead to demagnetization in strong ferromagnets via spin-flip scattering events,<sup>30,31</sup> where an electron from the majority spin fills the hole in the minority spin, as depicted in Fig. 1(a). This flipped spin could then decay into spin waves as illustrated in Fig. 1(b), which induces a band mirroring in the nearby atoms where the quantization axis has now changed. By using time-resolved XAS and XMCD, we aim at uncovering the different timescales and energies of the different processes involved.

Figure 2 shows transient XAS and XMCD of laser-induced holes below and above the Fermi level. XAS [Fig. 2(a)] and XMCD spectra [Fig. 2(b)] were measured at a fixed time delay of 0.4 ps at the Co  $L_3$  edge. The pump induced changes are shown as green symbols and shading. While the change in XMCD appears to be mostly an homogeneous reduction at all photon energies, the change in XAS clearly displays a derivative-like shape with a zero crossing at an x-ray energy of 777.2 eV (see top axis of Fig. 2) as indicated by the dashed vertical line. At lower x-ray energy, the XAS signal is increased as expected for fs laser-induced hole states. At higher energy, XAS transitions into previously unoccupied states are blocked by laser-excited electrons leading to the observed intensity reduction. It is, therefore, possible to identify 777.2 eV as the position of the Fermi level (see bottom axis of Fig. 2).<sup>41</sup>

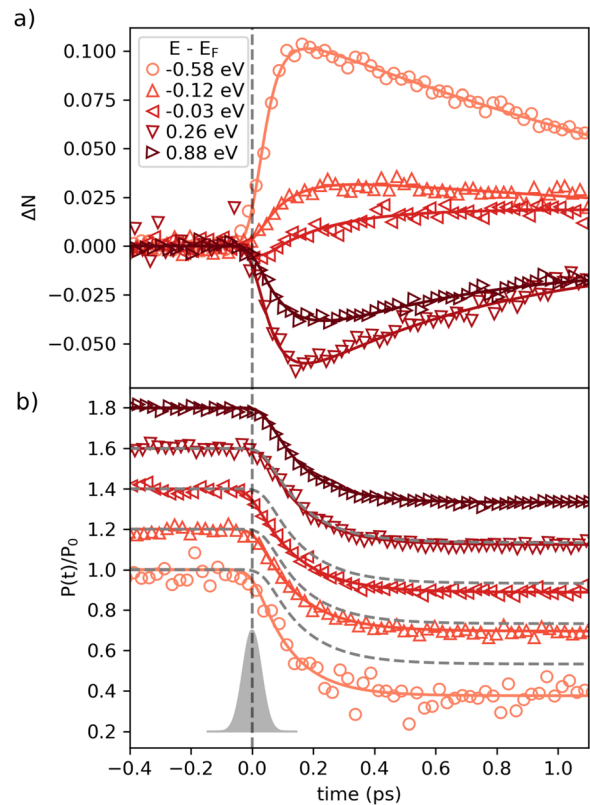


**FIG. 2.** Pumped, unpumped, and their differences in (a) XAS and (b) XMCD at the Co  $L_3$  edge at a delay of 0.4 ps. The vertical dashed line indicates the position of the observed zero crossing at the Fermi level  $E_F$ . The photon energy is shown on the top axis, while the energy with respect to the Fermi level is shown on the bottom. The differences (pumped–unpumped) are shown on a separate vertical axis on the right. For the XMCD difference, the sign was reversed to ease visual comparison with the unpumped XMCD profile.

Figure 3 displays time-delay traces obtained for various state energies relative to the Fermi level,  $E-E_F$ . Below  $E_F$ , the curves display initial increases in the XAS intensity followed by subsequent decays on timescales longer than several 100 fs. Above  $E_F$ , the transient XAS changes are negative while directly at  $E_F$  a more complex behavior emerges. In the following, we describe these observations in terms of changes in the hole population,  $\Delta N$ , at state of energy  $E-E_F$ . The small contribution due to spin-orbit coupling of the state-resolved XAS intensity<sup>42,43</sup> will be neglected here. It is important to emphasize that  $\Delta N$  can also include time dependent changes in the electronic structure.<sup>44</sup> It is apparent in Fig. 2(a) that such electronic structure changes indeed occur. For instance, at  $E-E_F$  near 4 eV, i.e., much higher than the pump photon energy, the observed variations of  $\Delta N$  are unlikely to be caused by the population dynamics of electrons in these states.

The curves for  $\Delta N$  in Fig. 3(a) were fitted with a double exponential to describe an excitation and a relaxation process. The fit parameters are summarized in Table II in the supplementary material. Far above and below the Fermi level, the initial rise times of  $\Delta N$  are essentially determined by the length of the pump pulses. The subsequent decay timescales are shorter further away from the Fermi level as one would expect from a Fermi liquid behavior of the electronic system.

The XMCD spectra can be described as being proportional to the product of state-dependent population,  $N$ , and a polarization term,  $P$ .

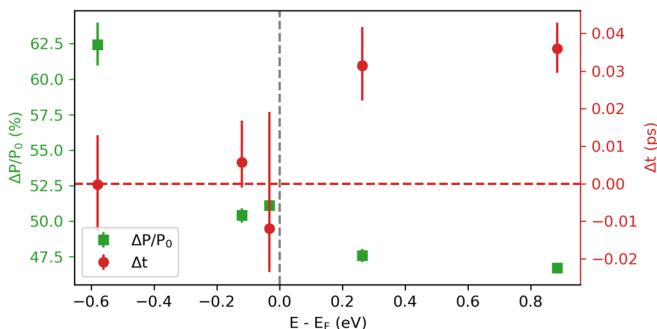


**FIG. 3.** Time-resolved change in-state resolved (a) charge  $\Delta N$  and (b) relative polarization change  $P(t)/P_0$  around the  $E_F$  at the  $L_3$  edge. In (b), the data are shifted vertically for clarity and the gray dashed curves are the fit as explained in the text at  $E-E_F = 0.88$  eV.



The latter contains both spin and orbital polarization with the orbital contribution being significantly smaller than the spin polarization as shown in the sum rule analysis detailed in the [supplementary material](#).<sup>42,43</sup> Similar to conventional sum-rule-analysis of time-resolved XMCD spectra, the magnetic dipole term can be neglected for our poly-crystalline samples<sup>45,46</sup> and was found to be negligible in similar samples.<sup>47</sup> Using the results for  $\Delta N$  from Fig. 3(a), we can separate the state polarization from state-dependent charge dynamics. The time-resolved polarization dynamics, normalized to the ground-state polarization of the respective states, are shown in Fig. 3(b). The individual experimental results (symbols) are shifted vertically for clarity and are compared to exponential polarization decays that include the magnitude of the decay,  $\Delta P$ , the decay time constant,  $\tau$ , and a delayed demagnetization onset,  $\Delta t$ , as fit parameters. All parameters are summarized in Table II in the [supplementary material](#). To highlight the difference between the curves below and above the Fermi level, the same relative polarization dynamics for  $E-E_F = 0.88$  eV are shown as a dashed gray curve together with each trace. This allows two visual observations. First, the amount of demagnetization is not the same at each value of  $E-E_F$ . Clearly, the demagnetization is significantly stronger below  $E_F$  than above. Second, there is a time delay,  $\Delta t$ , apparent in the response, with faster dynamics for states below  $E_F$ . The complete fitting model and analysis of the uncertainties on the fitted values are presented in Table II in the [supplementary material](#). In Fig. 4, the relative change in polarization  $\Delta P/P_0$  and the time lag  $\Delta t$  are shown as function of  $E-E_F$ . We stress here that this time lag  $\Delta t$  is determined by the delayed apparent response of the relative change in polarization  $\Delta P/P_0$  with respect to the charge dynamics  $\Delta N$ , for the same given photon energy. It can also be visualized by comparing the relative change in polarization  $\Delta P/P_0$  at different photon energy, but this is not how we extracted it.

The data that we presented in this Letter conclusively demonstrate a vastly different magnetization dynamics above and below the Fermi level for Co  $3d$  levels in [Co/Pd] multilayers. Below  $E_F$ , the ultrafast drop in magnetic polarization is up to 32% larger than above (see Fig. 4). This is clearly outside any experimental uncertainty as demonstrated in Fig. 3(b). Moreover, the onset of the polarization dynamics occurs simultaneously to the charge dynamics, i.e.,  $\Delta t = 0 \pm 10$  fs. This is the behavior expected for individual electrons/holes being scattered between different electronic states as depicted in Fig. 1(a). This also leads to Stoner excitations where electrons/holes are scattered between the spin-up and spin-down states.<sup>28</sup> In strong



**FIG. 4.** Fitted relative change in polarization  $\Delta P/P_0$  and time lag  $\Delta t$  in polarization change response as function of  $E-E_F$  at  $L_3$  edge.

ferromagnets, such as [Co/Pd] multilayers, spin-flip scattering can only occur for states below  $E_F$  where spin-up and spin-down states are hybridized via spin-orbit coupling.<sup>31</sup> The same Elliot-Yaffet-type spin-flip scattering processes are thought to also transfer spin angular momentum to the lattice.<sup>29-31</sup>

However, for ultrafast demagnetization to occur, the flipped spins of individual electrons/holes need to be transferred to the whole electronic system. This usually takes place via the formation of collective spin excitations, i.e., spin waves. In the Heisenberg model, spin waves lead to slight changes in the atomic spin quantization axis and result in a mixing of spin-up and spin-down states as observed in photoemission spectroscopy.<sup>48</sup> This situation is depicted in Fig. 1(b). Since the formation of spin waves takes time,<sup>34</sup> we expect a characteristic time delay relative to the instantaneous demagnetization of individual electrons/holes. We assign the observed delayed onset of demagnetization above  $E_F$  of  $\Delta t = 35 \pm 10$  fs (see Fig. 4) to this effect. This is observable above the Fermi level since there the majority of unoccupied states reflects the atomic magnetic moments.<sup>49,50</sup> We note that spin current from Co to Pd layers can occur on this timescale.<sup>51</sup> However, it remains unclear how such effect would delay rather than contribute to the demagnetization of the Co spin system above the Fermi level.

In summary, taking advantage of the high brightness available at free electron laser (FEL) and improved  $I_0$  normalization scheme, we were able to show that time-resolved XAS and XMCD spectroscopy can provide detailed information in the microscopic mechanism at play during ultrafast laser excitation. In particular, we report on different dynamics of the spin system below and above the Fermi level. This is manifested by both a 32% larger change in spin dynamics below the Fermi level and a  $35 \pm 10$  fs delayed response above it. Both of these effects suggest a scenario for a strong ferromagnet where spin-flips occur preferentially below the Fermi level where spin-up and spin-down states are hybridized. Moreover, we also report on initial evidence that indicates effects beyond a simple electronic redistribution and demagnetization with changes in XAS observed 4 eV above the Fermi level, suggestive of band structure dynamics. With ever improved normalization schemes and higher repetition rate FELs, transient near-edge soft x-ray spectroscopy promises to be a valuable tool in understanding out-of-equilibrium phenomena.

See the [supplementary material](#) for the sample preparation, sum-rules analysis, timing drift correction, pump and probe absorption profiles in the sample, and finally the complete fitting model with uncertainty estimation.

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## AUTHOR DECLARATIONS

### Conflict of Interest

The authors have no conflicts to disclose.

## DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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