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Supplementary Materials for

Ultrafast optically induced spin transfer in ferromagnetic alloys

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Supplementary Text

Section S1. Spectrum

In this section, we show an example of a reflected HHG spectrum from the FeNi sample. The spectrum is obtained by adding the recorded intensities for the two opposite magnetization directions (see fig. S1). Due to the discrete nature of the generated XUV photons (FWHM of a single harmonic ~ 0.8 eV here), the magnetic information available for photon energies close to the valleys is rather noisy and not suitable for a spectral analysis. The spectral resolution in the experiment is below 200 meV and is limited by the energy resolution of the spectrometer. This was determined by comparing the same high harmonics to a commercial spectrometer.

Section S2. Majority dynamics

Figure S2 shows the difference in the transient occupation in the *majority* channels of Ni and Fe. Therefore, it is the counterpart to Fig. 2 of the manuscript, which shows the *minority* channels. The scaling is identical to the scaling of Figure 2 in the manuscript in order to demonstrate that the OISTR effect is much smaller in the majority channels. Note, however, that the calculated magnetization dynamics of Fig. 3 in the manuscript contains the combined dynamics of minority and majority electrons.

Section S3. Two separated core levels for Ni

As discussed in the manuscript, the experimental TMOKE asymmetry at a given photon energy correlates to the spin-polarization of those states that are contributing to the optical transition, i.e. the initial and final state. In order to unambiguously extract information about the dynamics of the final states close to the Fermi-level, the initial states need to be spin-degenerate and

energetically sufficiently narrow. The core level energies of the Ni $M_{2,3}$ edges, however, are separated by 1.8 eV (66.2 eV and 68.0 eV, respectively; see yellow and red lines in fig. S3A). Therefore, the asymmetry of Ni is a superposition of two times a magnetooptical contrast stemming from the same final states, but shifted by 1.8 eV.

Nevertheless, this fact does not influence the interpretation obtained in the manuscript. First, we recall that the spin dynamics at the Fermi-level is probed at energies corresponding to the binding energies of the core levels, which is at 66.2 and 68.0 eV for the M_3 and M_2 absorption edge for Ni, respectively (see fig. S3A). We additionally note that the required signal-to-noise ratio in this analysis procedure did restrict us to energetic regions with high peak intensities of the XUV light (i.e. odd harmonics of the driving laser, see fig. S1). The two relevant traces that are presented for Ni in the manuscript are analyzed at photon energies of 64.2 eV (dark blue curve in Fig. 3B) and ~70 eV (dark blue curve in Fig. 3C). These energies correspond to -2 eV below the Fermi-level and 4 eV above the Fermi-level with respect to the M_3 absorption edge and - with respect to the M_2 edge - translate to -4 eV below the Fermi-level and 2 eV above the Fermi-level.

Let us now focus on the energy region below the Fermi-level. The colored shaded areas (M_2 red, M_3 yellow) in fig. S3B mark the energy regions relevant for OISTR (see e.g. calculations in Fig. 1C of the manuscript) in Ni with respect to their corresponding absorption edge. From this plot, we can deduce that there is only a small photon energy region between 65eV and 66 for which the spectroscopic OISTR signal of the M_2 edge overlaps with the OSTR signal of the M_3 absorption edge. Fortunately, in our spectroscopic analysis, it is straightforward to extract the spectroscopic OISTR signal from an energy region that is well separated from the overlapping

OISTR region of the M_2 and M_3 absorption edges. With an excitation (pump) energy of 1.5 eV there is little to no OISTR dynamics excited at energies -4 eV below the Fermi-level. Furthermore, the magnetic asymmetry rapidly decays in Ni below the Fermi level. Therefore, at 64.2 eV (dashed blue line in fig. S3B) we predominantly probe the asymmetry from states with transitions from the M_3 absorption edge (i.e. there is no overlap with the shaded red OISTR region). Accordingly, the OISTR dynamics shown in the dark blue curve in Fig. 3A of the manuscript is almost not disturbed by dynamics from states with transitions of the M_2 absorption edge and is indeed evolving on the timescale of the optical excitation.

A similar argument holds true for photon energies at 70 eV (corresponding to 2 eV and 4 eV above the Fermi-level, respectively): As supported by the TD DFT calculations, there is no OISTR signature to be expected at these energies. In fact, we do not see such a fingerprint in the experimental data (dark blue line in Fig. 3C of the manuscript). Additionally, the OISTR effect is prevailing on an earlier timescale than the subsequent conventional demagnetization. Therefore, these two processes can be separated - even in a superimposed signal. For Fe the energetical difference between the M_2 and M_3 absorption edge is smaller than 100 meV and can be neglected within our energy resolution. Hence, the above discussed ambiguity regarding the interpretation of the spectral analysis does not exist for Fe.

Section S4. Energy integrated signals

Figure S4 shows the integrated signal over the whole Fe and Ni absorption edges. Therefore, it is directly comparable to the work in (*34*). One immediately recognizes that the delay between the Fe and Ni traces is larger than for the case of the FeNi alloy investigated in (*34*). This finding is fully consistent with our observation of the increased OISTR effect in the present $Fe_{50}Ni_{50}$

sample, which leads to an increased offset between the two magnetic subsystems during the optical excitation. Afterwards in both cases the sub-lattice magnetization decays with the same rate for Fe and Ni due to exchange scattering processes.

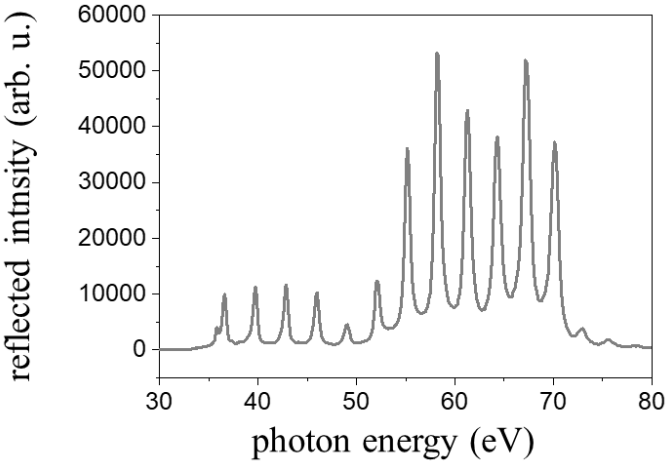

Fig. S1. High-harmonic spectrum reflected from the FeNi sample. The data was obtained by adding up the recorded reflected intensities for two opposite magnetization directions.

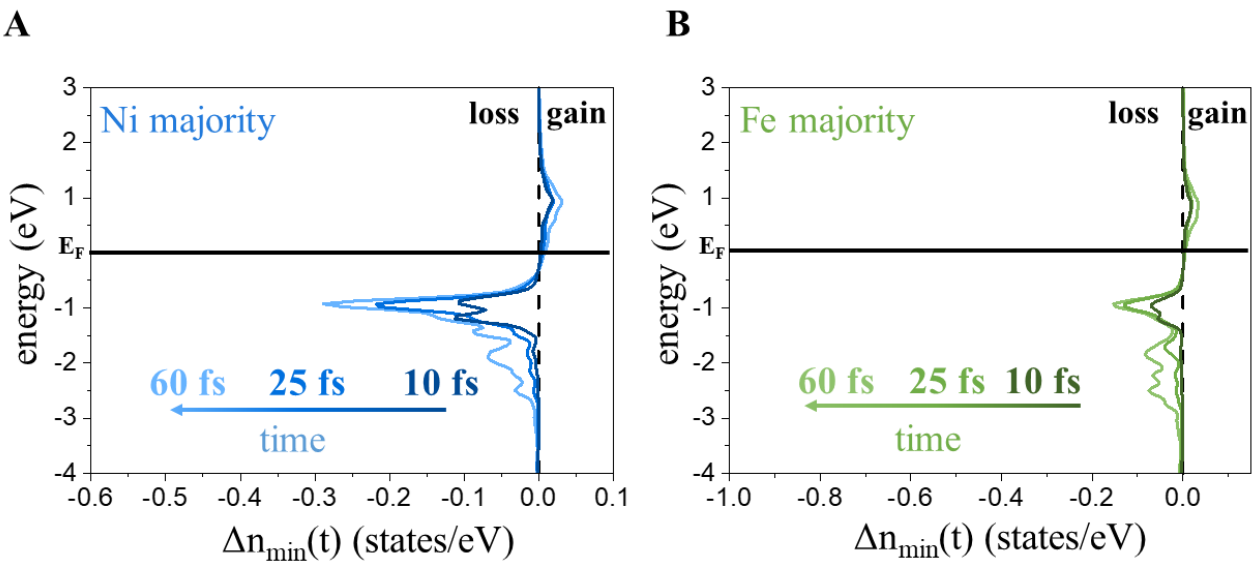

Fig. S2. Transient occupations in Fe and Ni majority channels. (**A** and **B**):Difference of the transient occupation compared to the unexcited case in the majority channels of Ni (A) and Fe (B) at characteristic time steps. The scaling was chosen according to Fig. 2 of the manuscript and demonstrates that the OISTR dynamics is much more pronounced in the minority part of the spectrum.

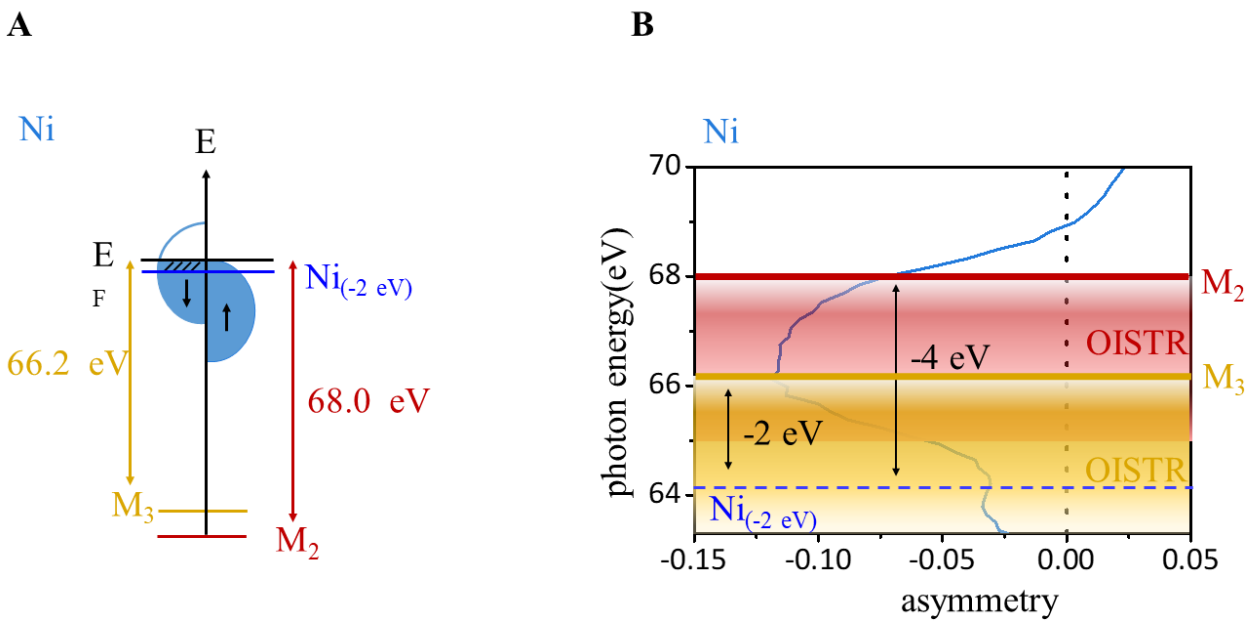

Fig. S3. Schematic depiction on the influence of two separated core levels for Ni. (**A**) The probing process with the HHG starting from the M2 and M3 absorption edges corresponds to a superposition of two times the same spin polarization of the final states, separated by 2 eV. However, this does not influence the interpretation regarding the dynamics of Ni in the manuscript. (**B**) The blue interrupted line marks the energy that is analyzed in Fig. 3B of the manuscript. This energy translates to -4 eV below the "M2 –Fermi-level" (brown line marked M2, see text). Fortunately, with an excitation energy of 1.5 eV there is little to no OISTR dynamics excited at this energy of -4 eV below the Fermi-level. Therefore, the dynamics labeled with "Ni (-2 eV)" in the manuscript directly serves as a fingerprint for the OISTR dynamics. In a similar argument, the dynamics probed in Fig. 3C of the manuscript is - with respect to both Fermi-levels - not affected by the OISTR dynamics.

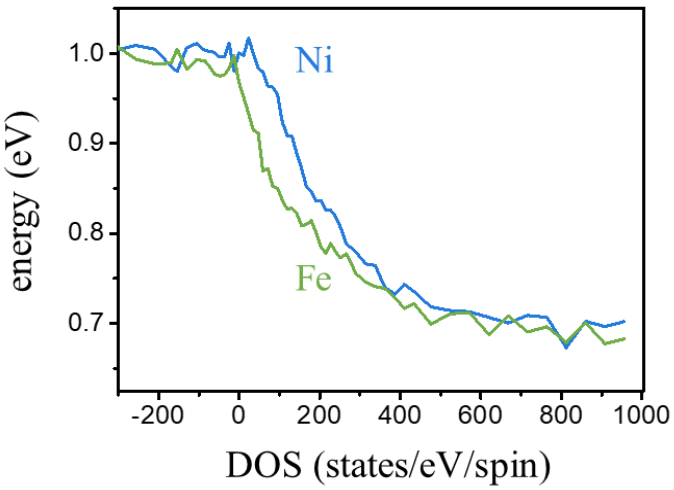

Fig. S4. Energy integrated magnetization dynamics for Fe and Ni in $Fe_{50}Ni_{50}$. The data reveals a visible initial delay of Ni compared to Fe caused by the pure spin transfer from Fe to Ni during the optical excitation (i.e. OISTR). Afterwards, this delay vanishes due to exchange scattering processes on a timescale of several hundred femtoseconds. Finally, subsequent phonon-mediated spin flips reduce the magnetization of both sublattices equally further.