The influence of the excitation pulse length on ultrafast magnetization dynamics in nickel

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The laser-induced demagnetization of a ferromagnet is caused by the temperature of the electron gas as well as the lattice temperature. For long excitation pulses, the two reservoirs are in thermal equilibrium. In contrast to a picosecond laser pulse, a femtosecond pulse causes a non-equilibrium between the electron gas and the lattice. By pump pulse length dependent optical measurements, we find that the magnetodynamics in Ni caused by a picosecond laser pulse can be reconstructed from the response to a femtosecond pulse. The mechanism responsible for demagnetization on the picosecond time scale is therefore contained in the femtosecond demagnetization experiment.

I. INTRODUCTION

For ultrafast demagnetization of a ferromagnet, spin angular momentum needs to be transported away from the spin system. Unlike other ultrafast processes in solids, the time scale of magneto-dynamics is believed to be dictated by the transfer of angular momentum. Therefore, the femtosecond magnetization dynamics is of fundamental interest. Laser-induced demagnetization is also of technological interest for magnetic recording devices: in order to optimize hard disks for fast writing times, long data retention times, and high storage densities, conventional writing needs to be combined with pulsed laser heating. The heat assisted magnetic recording (HAMR) technique therefore combines a magnetic field source with a pulsed laser diode to heat up the magnetic bit during writing. Although the desired effect of the laser pulse for HAMR lies in the softening of the magnetic bit, it leads to a reduction of the magnetic moment simultaneously, a process investigated in this experiment.

The demagnetization process is expected to be caused by both, the excited electron gas as well as the lattice. If the ferromagnet is heated by a femtosecond laser pulse, the electron gas initially reaches a temperature, which exceeds the lattice temperature. This initial temperature rise in the electron gas is followed by thermalization with the lattice to a common temperature. In case of excitation by a picosecond laser pulse, the electron gas and the lattice stay close to thermal equilibrium during the whole demagnetization process. For the two main models of ultrafast demagnetization, the Elliot-Yafet spin flip scattering model as well as the superdiffusion model, the excited electron gas is the driving force for the femtosecond magnetization dynamics. Additionally, in case of the Elliot-Yafet mechanism, the lattice temperature promotes the high spin flip probability necessary for demagnetization. It is likely that both processes, transport as well as bulk spin flips, play an important role for ultrafast demagnetization. The generated spin currents have been observed experimentally by several groups. At the same time, the model from Koopmans et al. is very successful in explaining the ultrafast loss of magnetization in a variety of materials.

Here, we investigate if the demagnetization caused by a picosecond laser pulse can be described by the same mechanism as the demagnetization by a femtosecond pulse. This is...
achieved by studying the ultrafast demagnetization dynamics of a Ni film as a function of the pump pulse length.

II. EXPERIMENTAL SETUP

We employ the magneto optical Kerr effect (MOKE) in longitudinal geometry to access the optically induced magneto dynamics of a 20 nm nickel film. The sample is grown by e-beam evaporation on a glass substrate. The layer structure is 5 nm Cr/50 nm Cu/20 nm Ni/3 nm Ti. The experiment is performed in a pump-probe setup\(^1\) (Fig. 1(a)). An amplified femtosecond laser system provides 800 nm pulses with a repetition rate of 10 kHz. Individual pulse compressors are used for the pump- and probe pulses in order to perform pulse length dependent measurements. The probe branch compressor (CP 2) is adjusted to deliver pulses as short as possible (40 fs full width half maximum (FWHM) of the autocorrelation trace, fit to a Lorentzian pulse shape, corresponding to a FWHM pulse width of \(\approx 20\) fs). In contrast, the pump branch compressor (CP 1) is used to deliberately change the pump pulses’ length from approx. 40 fs–7 ps FWHM auto-correlation width.

The MOKE is measured by frequency doubled probe pulses (400 nm) to circumvent state blocking effects.\(^{16,17}\)

The pulse energy of the differently long pump pulses is kept constant at \((88 \pm 1) \mu J\). This results in a pump energy density of 1.5 mJ/cm\(^2\) per pulse on the sample. Notice that this is the incident pump fluence: A significant fraction of the pulse is reflected or absorbed in the Ti cap layer. During the pump-probe measurements, an external in-plane magnetic field of \(H = \pm 15\) mT is applied to keep the sample in a \((\uparrow, \downarrow)\) fully saturated state \((M_{\uparrow,\downarrow})\). Lock-in detection is used to measure the pump induced change of the polarization rotation \((\Delta \theta_{\uparrow,\downarrow}(t))\) by a balanced photodiode-detector. The pump beam is mechanically chopped at a frequency of 83 Hz. In addition, a \(\lambda/4\)-plate is used before the balanced detector to suppress spurious elliptic contributions. The pump induced change of the magnetization is determined as

\[
\Delta M_{\uparrow,\downarrow}(t) = \frac{1}{H} \frac{\partial H}{\partial t} \Delta \theta_{\uparrow,\downarrow}(t)
\]

FIG. 1. (a) Schematic representation of the laser setup for the Kerr measurements: Two pulse compressors (CP 1,2) allow to adjust the pulse length of the pump and the probe beams independently. A 400 nm band pass filter (BPF) suppresses any 800 nm contribution of entering the detector. A \(\lambda/4\)-wave plate is used to eliminate elliptic contributions. (b) The cross correlation setup used to measure the pulse shape of the pump pulses. The pump pulse is up-converted by a 20 fs, 800 nm pulse to 400 nm where it is detected by a photo diode.
\[
\frac{\Delta M(t)}{M_0} = \frac{\Delta \theta_{\parallel}(t) - \Delta \theta_{\perp}(t)}{\theta_{\parallel} - \theta_{\perp}},
\]

(1)

Here, \(\theta_{\parallel,\perp}\) represents the static MOKE rotation. Notice that Eq. (1) can only be valid, if the relative pump induced reflectivity change is significantly smaller than \(\Delta M(t)/M_0\). This condition has been verified experimentally.

The pump pulses’ length is first measured with an autocorrelator. These traces are shown in the inset of Fig. 2. Each curve is fitted by a Gaussian function. The FWHM of these fits is used to label the pump pulse lengths throughout this paper (legend in Fig. 2). As the autocorrelation of any signal is always symmetric, the measured autocorrelation does not directly represent the temporal shape of the pump pulses. To overcome this limitation, we use the cross-correlation technique to measure their actual temporal intensity profile (Fig. 1(b)). For that the pump pulses of variable length are overlapped with 800 nm sampling pulses of \(\approx 20\) fs length in a Beta-Barium-Borate (BBO) crystal. The intensity of the generated 400 nm cross-correlation signal is measured by a photodiode. The result is depicted in Fig. 2 and represents the temporal shape of the pump pulses. The setup allows only for an angle of 5° between the incoming beams which resulted in a temporal broadening of the cross-correlation signal of about 100 fs. Due to this effect, the cross-correlation of the 40 fs trace in Fig. 2 appears significantly broadened.

III. RESULTS

Figure 3(a) shows the relative change of the pump induced demagnetization for various pulse lengths. Pump pulses with a length below 1 ps show an ultrafast demagnetization followed by an ultrafast (and subsequent slow) remagnetization. Longer pulses, however, show only the slow remagnetization. Surprisingly, all measured MOKE traces match at delays longer than \(\approx 10\) ps. This shows that the demagnetization after 10 ps only depends on total energy of the pulse, not on the pulse shape. Thus, the long time remagnetization dynamics is solely governed by a common temperature of the electron, spin, and lattice system provided by the totally absorbed pulse energy.

The time at which demagnetization reaches a minimum scales linearly on pump pulse length (blue squares) depicted in Fig. 3(b). The maximum demagnetization of 5% is reached for the shortest pulse of 40 fs. However, the maximum demagnetization depends crucially on the length of the used pump pulses. The shorter the pump pulses, the larger the achieved demagnetization. It is evident that the demagnetization is governed by two regimes: for pump pulses with a pulse length \(< 2\) ps, the demagnetization depends much stronger on the pulse length than for pump pulses \(> 2\) ps. This effect can be quantified by the slopes of the demagnetization amplitudes for the two regimes (Fig. 3(b)). The slope \(m_{<2\text{ps}} = -1.5\% / \text{ps}\) and \(m_{>2\text{ps}} = -0.2\% / \text{ps}\). They differ at least a factor \(m_{>2\text{ps}} / m_{<2\text{ps}} \approx 7\) from each other. The crossover of the two regimes happens at approx. 2 ps, corresponding to the electron-phonon equilibration time. At 2 ps, the demagnetization of 3% corresponds to a temperature increase of 50 K.

![FIG. 2. Pump pulse length measured with the cross-correlator (described in the text) and an autocorrelator (inset).](image-url)
Our experiment demonstrates that a laser pulse shorter than 2 ps causes two components of $\Delta M = \Delta M_f + \Delta M_s$, distinguished by the recovery time: There is a component, which recovers within 2 ps (named $\Delta M_f$). A second (slow) component, named $\Delta M_s$, recovers on a time scale of 50 ps (1/e-decay time). For excitations with pulses longer than 2 ps, only $\Delta M_s$ remains and its amplitude is weakly dependent of the pulse length (visible in Fig. 3 for delay times $>4$ ps).

The question arises if the demagnetization process caused by a picosecond pump pulse can be described by the same processes as the demagnetization by a femtosecond pulse. If the

![Diagram](image-url)

**FIG. 3.** (a) Relative demagnetization for various pump pulse lengths. (b) Maximum demagnetization and time of the magnetization minimum plotted versus pump pulse length.

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**FIG. 4.** Measured demagnetization from Fig. 3(a) (circles) compared with the calculated demagnetization using Eq. (2) (lines). This result indicates that the response to a picosecond pump pulse can be reconstructed from the response to a femtosecond pump pulse and the picosecond pump pulse shape.
magnetization reacts in such a linear way to the excitation, one can construct the response for any pump pulse \( p(t) \) by convolution with the response to a \( \delta \) pulse as

\[
M\{p\}(t) = (M\{\delta\} * p)(t).
\]

Here, \( M\{p\} \) is the linear response of the magnetization to a stimulus \( p \), \( \delta \) is the delta-distribution, i.e., an ultra short excitation. In our measurement, the shortest excitation pulse is 40 fs. This is significantly shorter than the longest pulse of 7 ps. In Fig. 4, we show the convolution of the magnetization dynamics for the 40 fs pulse with the shape of the longer pulses \( p(t) \). The calculated response is compared to the Kerr measurements. The convolution reproduces the measured demagnetization curve. The time offset of the convolution as well as the amplitude have been adjusted to match the measured response. This demonstrates that the demagnetization dynamics can be reconstructed by the measurement of a single ultrafast demagnetization trace for the level of excitation used in this experiment.

IV. CONCLUSIONS

Our experiment demonstrates that the demagnetization by a picosecond laser pulse is described by the response to a femtosecond pulse convoluted with the pulse shape of the picosecond pulse. Therefore, the dynamics driven by the femtosecond pulse contains all information about the response to a picosecond pulse and is driven by the same physical processes. We demonstrate this for demagnetization amplitudes of up to 5%. The demagnetization shows two components. One of them is independent of the pump pulse length and recovers on a time scale of heat diffusion. The second component is only present for pump pulses of less than 2 ps length. We interpret the decay of the fast component to be caused by the fast cooling of the electron gas. The fast demagnetization contribution can be achieved efficiently and quickly, yet, it is not a long lasting effect.

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