Electronic excitations in ferromagnets are essentially spin-polarized. In the transition metal ferromagnets Fe, Co, and Ni excitations of 0.1–1 eV relax on femtosecond to picosecond time scales \cite{ref1, ref2} due to scattering with secondary excitations mediated by e-phonon, e-magnon, and e-e/ exchange interaction. The underlying microscopic processes are essential in ultrafast magnetization dynamics \cite{ref3, ref4}.

Recently, this field has been propelled by ultrafast spin-polarized \cite{ref5, ref6} and unpolarized currents \cite{ref7} generated by a gradient in excitation density \cite{ref8}. These effects offer to exploit non-local magnetization dynamics. Already now femtosecond (fs) laser-excited spin currents in layer stacks were reported to induce spin transfer torque \cite{ref9}. Furthermore, laser-induced spin currents in a magnetic tunnel junction were controlled by a bias voltage \cite{ref10}. Moreover, spin currents are reported to drive an ultrafast change between ferri- and ferromagnetic order \cite{ref11}.

Such demonstrations suggest an extension of spintronics, including spin filter and magnetoresistance effects, into the non-equilibrium regime. In turn, fs time-resolved experiments allow conclusions on spin transport \cite{ref12, ref13, ref14, ref15, ref16}, to shed light on the underlying microscopic mechanisms. However, up to date experimental means to distinguish ultrafast spin currents and competing depolarizing spin-flip scattering, which reduces the magnetization \( M \) in spin current sources like ferromagnetic layers, are missing.

In this Letter, we show how to fill this gap by establishing that laser-excited spin currents result in ferromagnetic Co/Cu(001) films in a specific transient magnetization profile \( M(z,t) \) in the direction \( z \) along the film normal, before spin flips impact the dynamics. Key is the combined analysis of the film thickness dependence with the time-dependent difference in polarization rotation \( \theta \) and ellipticity \( \epsilon \) of the magneto-optical Kerr effect (MOKE). We separate spin currents generated by non-thermalized carriers up to \( \sim 100 \) fs and local demagnetization due to spin-flip scattering of thermalized carriers dominant after \( \sim 200 \) fs.

A contested issue in ultrafast MOKE is whether the observations reflect \( M(t) \), or transient changes of the optical constants. Koopmans et al. \cite{ref17}, finding that \( \theta \) and \( \epsilon \) of the complex Kerr angle \( \Phi = \theta + i \epsilon \) showed a different behavior for \( t \leq 100 \) fs, assigned the difference to statefilling effects. Later, Guidoni et al. \cite{ref18} reported that the MO response is dominated by the magnetization dynamics after electron thermalization. However, the origin of the transient difference between \( \theta \) and \( \epsilon \) before thermalization is still unclear \cite{ref19}.

Also, the relative importance of (i) spin-flip scattering of thermalized electrons, described by the microscopic three temperature model \( (M3TM) \) \cite{ref20}, and (ii) superdiffusive spin transport \( \sim 200 \) fs, while demagnetization of metallic films on insulators was ascribed to spin flips \cite{ref21}, leading to an emerging consensus that both processes play a role \cite{ref22}.

In Fig. 1(a) we contrast the relative local magnetization \( m(z,t) = M(z,t)/M_0 \) for (i) spin-flip scattering of thermalized electrons and (ii) spin transport, calculated with an extended M3TM and a spin diffusion equation, respectively. \( M_0 \) is the homogeneous equilibrium magnetization. We calculated the time- and spatially dependent electronic and lattice temperatures, \( T_{e}(z,t) \) and \( T_{l}(z,t) \), respectively, for different Co film thicknesses \( d \) on Cu(001) employing a two temperature model \( (2TM) \) \cite{ref23, ref24}. Subsequently, we used the M3TM to obtain \( dm_{M3TM}/dt \) and \( m_{M3TM}(z,t) \) \cite{ref25, ref26}. To simulate spin diffusion (SD) we employ a diffusion equation for excited electrons based on Fick's second law for majority and minority electrons. Eq. 1 contains source, diffusion and decay terms. We use the diffusion coefficient \( D_s = v_s \lambda_s / 3 \)
the respective velocities and lifetimes \(2, 24\). We included electrons. The excitation probability follows the joint spin dependent mean free path \(\lambda\) according to the optical penetration length generated secondary electrons. To account for the spatial dependence of the magnetization profile are measured in a pump-probe experiment. (a) Normalized magnetization profile \(m(z)\) calculated with the M3TM \(17\) (solid line) for \(t_0 = 170\) fs and spin diffusion (dashed line) for \(t_0 = 20\) fs. (b) MOKE sensitivity \(\zeta(z)\) for the real part/rotation \(\theta\) (solid line) and imaginary part/ellipticity \(\epsilon\) (dashed line).

for self diffusion to simulate the transport, \(\sigma\) represents majority and minority electrons and \(\lambda_{\sigma} = v_{\sigma}\tau_{\sigma}\) is their mean free path along \(z\). The lifetimes \(\tau_{\sigma}\) and the electron velocity perpendicular to the surface \(v_{\sigma}\) are averaged up to 0.5 eV above the Fermi level \(22\). We obtain

\[
\frac{\partial n_{\sigma}}{\partial t} = D_{\sigma} \cdot \frac{\partial^2 n_{\sigma}}{\partial z^2} + c_{\sigma} \cdot S_{\sigma}(z, t) - \frac{n_{\sigma}}{\tau_{\sigma}},
\]

where \(n_{\sigma}\) is the density of excited electrons and \(S_{\sigma}(z, t)\) describes fs laser excited primary and subsequently generated secondary electrons. To account for the spatial distribution, we convoluted two exponential decay terms according to the optical penetration length \(\delta_{\text{kin}}\) and the spin dependent mean free path \(\lambda_{\text{ball}, \sigma}\) of primary excited electrons. The excitation probability follows the joint density of states \(23\). Then, \(\lambda_{\text{ball}, \sigma}\) was calculated with the respective velocities and lifetimes \(2, 24\). We included propagation in Cu with \(\lambda = 70\) nm \(23\) by setting \(n_{\sigma} = 0\), i.e. Cu acts as an electron drain. The last term \(-\frac{n_{\sigma}}{\tau_{\sigma}}\) describes electron thermalization and spin current decay by a time constant \(\tau_{\sigma}\). The change \(\frac{\partial m_{\text{SD}}}{\partial t}\) is defined through the balance of the magnetic moments of in- and outgoing electrons. As depicted in Fig. 1(a), \(m(z, t_0)\) are strikingly and systematically different for the two scenarios \(23\). While consideration of thermalized electrons results in a minimum \(m\) at the surface and a maximum at the Co-Cu interface, the spin diffusion description leads to a depletion of \(m\) at the interface in combination with a weak variation near the surface, in good agreement with a more sophisticated description \(8, 18\).

To analyze \(m(z, t)\) experimentally, we measured the complex MOKE \(\Phi = \theta + i \cdot \epsilon\) in a pump-probe experiment, illustrated in Fig. 1(top). The magneto-optical (MO) Kerr rotation \(\theta\) and the ellipticity \(\epsilon\) were determined by a polarization analysis using a balance detection scheme. We employed a cavity dumped Ti:sapphire oscillator which generates p-polarized 35 fs laser pulses at \(h\nu = 1.55\) eV and 40 nJ pulse energy at 2.53 MHz repetition rate. The incident pump fluence was 6 mJ/cm\(^2\). Epitaxial Co films of 4 nm \(< d < 20\) nm, which we investigated in situ, were grown in ultrahigh vacuum on Cu(001) following Ref. \(20\). The film’s easy axis of magnetization lies in the film plane and we measure MOKE in the longitudinal geometry.

We find different transient responses of \(\theta(t)\) and \(\epsilon(t)\), shown in Fig. 2. Our following conclusions are based on the fact that Kerr rotation \(\theta\) and ellipticity \(\epsilon\) exhibit different depth sensitivities which we use to identify specific magnetization profiles \(m(z, t)\) caused by spin-flip scattering of thermalized electrons or spin polarized currents, see Fig. 1(a). In Fig. 1(b) we plot \(\zeta = \zeta_0 + i \cdot \zeta_e\), which we term MOKE sensitivity for the complex Kerr angle \(\Phi = \theta + i \cdot \epsilon\), as a function of the position \(z\) within a 20 nm thick film. The sensitivity \(\zeta\) indicates how changes \(\Delta m(z, t) = m(z, t) - 1\) contribute at a depth \(z\) in first order to the total change \(\Delta \theta(t)/\theta_0 + i \cdot \Delta \epsilon(t)/\epsilon_0 = \int_0^z \zeta(z) \cdot \Delta m(z, t) dz\).

Following Traeger et al. \(27\), \(\zeta(z)\) was calculated by setting \(m = 0\) with the exception of a part \(z, z + dz\) where \(m = 1\), and determining \(\Phi_{dz} = \theta_{dz} + i \cdot \epsilon_{dz}\) for this system. To take pump-induced changes into account we normalized \(\phi_{dz}\) to the equilibrium values and obtain

\[
\zeta(z) \cdot dz = \frac{\theta_{dz}}{\theta_0} + i \cdot \frac{\epsilon_{dz}}{\epsilon_0}
\]

We used the matrix formalism by Zak et al. \(28\) with \(dz = 0.2\) nm and refractive indices \(n_{Cu} = 2.56 + i \cdot 4.26\) and \(n_{Co} = 2.56 + i \cdot 4.92\). The MO constant \(q_{Co} = 0.017 - i \cdot 0.020\) was determined experimentally.

Note that \(\zeta_0\) is larger near the surface than near the Co-Cu interface and \(\zeta_e\) exhibits a weaker \(z\)-dependence, see...
stronger reduction of $\epsilon$ compared to $\theta$ is not observed, likely due to a limited pulse duration and a remainder of the coherent artifact, in combination with a weaker $dm/dz$, see below. Changes for such $d$ are larger in $\theta$ than in $\epsilon$.

From the observed $\theta(t)$ and $\epsilon(t)$ we conclude on $m(z,t)$ and relate this to a spin-polarized current or spin-flip scattering, see Fig. 1(a). Here, the different depth sensitivity of $\theta$ and $\epsilon$, see Fig. 1(b), is essential. Remember, $\epsilon$ exhibits a stronger sensitivity at the Co/Cu interface than $\theta$. The stronger reduction of the ellipticity compared to the rotation starting within the pulse duration, see Fig. 2 thus implies that the film is demagnetized more near the inner interface than near the surface. The respective magnetization profile agrees qualitatively with the one obtained for SD, see Fig. 1(a). After some tens of femtoseconds, depending on $d$, $m(z,t)$ is concluded to be redistributed towards a profile expected from the M3TM \[17\], as can be seen in the larger variation of $\theta$ compared to $\epsilon$ in Fig. 2 at $t > t^*$, taking into account that $\theta$ has a higher sensitivity at the surface than $\epsilon$, see Fig. 1(b). These transient changes of the magnetization profile suggest that when the electronic system has not yet thermalized, a spin-polarized current dominates $m(z,t)$. With electron thermalization, spin-flip events take over. This is consistent with both the superdiffusive transport model \[6\] and the M3TM \[17\]. The superdiffusive transport model predicts spin transport for a non-thermalized electron system, which recedes with thermalization \[31\]. In the M3TM only thermalized electrons, which contribute to spin-flip scattering, are considered \[17\].

Our conclusions are corroborated by finding that the time delay $\tau_{\text{min}}$ of the minimum MO response exhibits a systematic thickness dependence, as depicted in Fig. 2. We performed a fitting analysis and determined $\tau_{\text{min}}$ using $f(t) = A_M \cdot (1 - \exp(-t/\tau_M)) + A_R \cdot (1 - \exp(-t/\tau_R)) + A_R' \cdot (1 - \exp(-t/\tau_R'))$ where $A$ are the amplitudes and time constants, respectively, $M, R, R'$ refer to demagnetization and recovery \[24\]. For $\epsilon$ and $\theta$ the minimum positions shift to later times with increasing $d$, see Fig. 3. Particularly interesting is the different thickness dependence of $\epsilon$ and $\theta$. By fitting the minimum’s position as a function of $d$ with a line, we find with slopes of $b_\epsilon = 13$ fs/nm and $b_\theta = 4$ fs/nm a pronounced, three time difference.

We simulated the thickness dependent $m(z,t)$ with the extended M3TM \[17\] and the spin polarized diffusion equation by adding both contributions

$$\frac{dm}{dt} = \frac{\partial m_{\text{M3TM}}}{\partial t} + \frac{\partial m_{\text{SD}}}{\partial t}. \tag{4}\text{.}$$

We calculated the MO response with eq. 4 and determined the delays of minimum MO response, which are included in Fig. 3. The overall shift of $\tau_{\text{min}}$ to later times with increasing $d$ is explained by the lower heat conductivity of Co compared to Cu \[31\]. Essential for the discussion of the competing spin-dependent processes is

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**Fig. 2.** Time-dependent relative MO responses of Kerr rotation $\Delta \theta(t)/\theta_0$ (dots) and ellipticity $\Delta \epsilon(t)/\epsilon_0$ (diamonds) of Co/Cu(001) for $d = 6, 10, 20$ nm. Solid lines represent analytical fit functions and guide the eye. For determination of both curves within 200 fs. $\theta$ probes effectively the near surface part, while $\epsilon$ rather averages over the film and includes a sensitivity at the Co/Cu interface. This depth dependence of $\zeta$ provides us with a probe for $m(z,t)$.

Now we consider our experimental results in detail. Fig. 2 depicts the time dependent relative MO responses $\Delta \theta(t)/\theta_0$ and $\Delta \epsilon(t)/\epsilon_0$ for Co/Cu(001) for $d = 6, 10, 20$ nm. The curves show a reduction of the signal which begins within the laser pulse and a subsequent recovery starting between 100 and 300 fs, depending on film thickness. For all $d$ we find differences for $\theta(t)$ and $\epsilon(t)$. For $d \geq 10$ nm we observe a faster reduction of $\epsilon$ than for $\theta$ and a crossing at delay $t^*$ of both curves within 200 fs. After this crossing the changes are inverted relative to each other and $\Delta \theta(t)/\theta_0$ remains stronger than $\Delta \epsilon(t)/\epsilon_0$ up to 25 ps (not shown). For $d < 10$ nm the initially
FIG. 3. (a) Time delay $\tau_{\text{min}}$ of minimal response in $\Delta \vartheta(t)/\vartheta_0$ (black dots) and $\Delta \epsilon(t)/\epsilon_0$ (red diamonds) vs. $d$. Solid lines are linear fits, dashed lines depict simulations. (b,c) Simulated $\Delta \vartheta(t)/\vartheta_0$ (solid) and $\Delta \epsilon(t)/\epsilon_0$ (dashed) for $d = 6$, 20 nm.

the thickness-dependent difference between $\tau_{\text{min}}$ for $\vartheta$ and $\epsilon$, which is qualitatively reproduced by our simulation. The interface region preferentially probed by $\epsilon$ reaches the minimum magnetization earlier and also starts to recover earlier. Consequently, this confirms our above explanation of the initial dynamics by spin transport near the Co/Cu interface and the later dynamics by spin-flip scattering at the surface.

We qualitatively reproduce the transient behavior of our observables already with a relatively simple model. In Fig. 3(b,c) we show two simulations for $d = 6$, 20 nm. For 20 nm we obtain an initially faster ($<100$ fs) change of $\epsilon$ compared to $\vartheta$, and later at $200$ fs the crossing of both curves. This behavior agrees with spin-dependent transport and the experimental observation in Fig. 2. For $t > t^*$, the pump induced variation for $\vartheta$ is stronger than for $\epsilon$, also in agreement with Fig. 2 and the dynamics are dominated by spin-flip scattering. This behavior is only obtained if the SD contribution is included, the M3TM alone is not sufficient. We conclude furthermore that the contributions from both mechanisms are comparable and therefore both have to be considered. However, as we demonstrate here, they can be separated in the time domain. For $d = 6$ nm we lose the sensitivity to the competing processes in the simulation, probably due to a more homogeneously demagnetized film. After all, Fig. 2(a) indicates a loss of SD at the inner interface as well as spin-flip scattering at the surface both on a length scale comparable to 6 nm. The deviation of the simulation and the experimental data in Fig. 3 is potentially a result of uncertainties in the optical constants. Moreover, for a quantitative description energy and spin dependent transmission coefficients at the interface need to be considered, as well as many body renormalizations, which is, however, beyond the scope of this letter.

A pump-induced variation of the optical constants due to the hot electron distribution could influence our results, but we are convinced that $m(z,t)$ dominates the dynamics. We argue as follows. (i) For films $d \geq 10$ nm the difference between $\vartheta$ and $\epsilon$ up to $t = 200$ fs is comparable to later times, but with opposite sign (Fig. 2). To explain this by a time dependent change of optical constants, a change in sign at a remaining absolute value would be required. This is rather unlikely since such effects would decay monotonously with the hot electron distribution within $\sim 1$ ps. (ii) With decreasing $d$ the difference between $\vartheta$ and $\epsilon$ at $t < t^*$ shrinks faster than for $t > t^*$ (Fig. 2), while the hot electron distribution remains for all $d$. In contrast, spin polarized transport is strongly affected by the sample thickness. (iii) The difference between $\vartheta$ and $\epsilon$ remains up to 25 ps, which is too long for state filling effects due to a hot electron distribution, considered previously.

In conclusion, we showed that the different depth sensitivity of Kerr rotation and ellipticity can be used to identify spatial magnetization profiles on ultrafast time scales resulting from spin polarized transport or spin-flip scattering. We found that the laser-excited spin dynamics in Co/Cu(001) films are dominated by spin transport effects on times up to $\sim 100$ fs when the electronic system has not yet thermalized, and by spin-flip scattering of thermalized electrons subsequently.

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