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Investigating the contribution of superdiffusive transport to ultrafast demagnetization of ferromagnetic thin films

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We experimentally investigate to what extent superdiffusive spin transport contributes to the ultrafast demagnetization of ferromagnets by pumping Ni thin films from the front side and back side of the sample. Within the experimental accuracy, the temporal evolution of the magnetization for front-side and back-side pumping is identical, hence no influence of transport was detected. Furthermore, adding a conducting buffer layer to enhance spin transport does not alter the magnetization dynamics significantly either. In order to explain the experimental results, angular momentum has to be locally dissipated on femtosecond timescales, supporting heating of the spin system as the main driving force for ultrafast demagnetization in the investigated ferromagnetic films. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4812658]

Since the observation by Beaurepaire et al. that a femtosecond laser pulse can quench the magnetization in the 3d-ferromagnets on sub-picosecond timescales, laser-induced magnetization dynamics has received a growing attention due to the prospect of ultrafast manipulation of magnetic order. Besides the importance for technological applications, research in the field has been motivated by the scientific interest in the microscopic processes governing the magnetization dynamics far from equilibrium and on unprecedented timescales. However, up until now no consensus on the microscopic origin of ultrafast demagnetization has been reached.

One of the most prominent explanations for laser-induced ultrafast demagnetization has, since its first observation, been heating of the spin system due to scattering of various (quasi-)particles, such as electron-electron, electron-phonon, or electron-magnon scattering. In this explanation, the electron system is initially heated by absorption of the laser pulse, whereafter scattering of hot electrons thermalizes the spin system. This means that angular momentum is locally transferred from the spin system to other systems, like, for example, the lattice. Ultrafast demagnetization in many experiments has been attributed to such a local dissipation of angular momentum.6,7

Recently, however, a completely different explanation for ultrafast demagnetization was proposed by Battiato et al., who calculated the demagnetization rate of Ni due to superdiffusive transport of majority and minority electrons after pulsed laser excitation. The majority spins move away from the excited region, because they have a longer mean free path than the minority electrons. As the probed region is the same as the excited region in a pump-probe experiment, this leads to an apparent demagnetization. It was claimed that superdiffusive transport could wholly explain the demagnetization process during the first few hundreds of femtoseconds in Ni thin films. The fact that spin transport plays a measurable role in ultrafast demagnetization has recently been shown by experiments on magnetic multilayers and systems with a network of magnetic domain walls. This begs the question whether indeed spin transport can fully explain the observed demagnetization in thin simple ferromagnetic films, as suggested by Battiato et al. Answering this question will prove to be of crucial importance for the interpretation of many experiments on femtosecond demagnetization reported in the literature.

In this letter, we perform a simple experiment to distinguish between the contributions of heating, i.e., local dissipation of angular momentum, and spin transport to the ultrafast demagnetization of a Ni thin film. The basic concept of our experiments is depicted in Fig. 1. The demagnetization of a Ni film on an insulating sapphire substrate is measured while exciting the film with a laser pulse from the front and from the back of the sample. In case of heating, the magnetization of the film is decreased throughout the whole film, hence in both the front-pump and back-pump scenario, a demagnetization is expected to occur. In case of transport, when pumping from the front, a demagnetization should be observed, as majority spins move away from the probe spot. However, when pumping from the back, these majority spins travel toward the probe spot, effectively increasing the probed magnetization. This means that comparing front-pump and back-pump experiments allows us to unambiguously pinpoint the main contributor to ultrafast demagnetization.

The ultrafast magnetization dynamics are measured by the Time-Resolved Magneto-Optical Kerr-Effect (TRMOKE) using a non-amplified Tsunami Ti:sapphire laser with a repetition rate of 80 MHz and a pulse length characterized by a FWHM of 75 fs. Before focussing the light on the substrate to a spotsize of \( \approx 8 \mu m \), the laser pulse power was \( \approx 0.5 nJ \), which assures that measurements are performed in the small fluence regime where contributions due to superdiffusive spin transport are not yet saturated. The Ni films are grown by DC magnetron sputtering on a sapphire substrate, which is transparent for the used 780 nm laser pulses but fully blocks electron transport due to the bandgap of 10 eV. No capping layer is used, thus a native oxide layer caps the sample. The Ni thickness \( d_{Ni} \) is wedged between 0 and 30 nm by a masking technique.

Demagnetization traces for front-pump and back-pump configurations are depicted in Figs. 2(a) and 2(b),
respectively. In both scenarios, a rapid demagnetization on the timescale of \( \approx 100 \) fs is observed for all \( d_{\text{Ni}} \), followed by a slower remagnetization on a picosecond timescale. This behavior is identical to earlier measurements on Ni films for small laser fluences while probing and pumping from the front.\(^{1,5,7}\) More importantly, the fact that for all Ni thicknesses a demagnetization is observed in the back-pump scenario means that contributions due to superdiffusive transport have to be smaller than thermal effects in all our measurements.

To see whether there are any traces of spin transport in the measurements, the demagnetization curves in Figs. 2(a) and 2(b) are investigated in more detail. A quick qualitative inspection of the raw data indicates that there is no substantial difference between both de- and remagnetization rates and demagnetization amplitude. For a more quantitative analysis, the data is fitted by a phenomenological fit function, and the resulting fits are plotted as lines in Figs. 2(a) and 2(b). After deconvolution of the fitted traces, the timescale for demagnetization \( \tau_m \) can be obtained by determining the delay time where the magnetization reaches \( 1 - 1/e \) times its maximum quenching. The insets of Figs. 2(a) and 2(b) display \( \tau_m \) as a function of \( d_{\text{Ni}} \) for front and back pumping, respectively. Both show no significant dependence on \( d_{\text{Ni}} \). Averaging \( \tau_m \) over the measured thicknesses yields \( \tau_m = (61 \pm 4) \) fs for the front pump configuration and \( \tau_m = (64 \pm 2) \), hence there is no significant difference between the two scenarios.

For both configurations, a clear decrease of the maximum magneto-optical quenching \( q_{\text{max}} \) is observed on increasing \( d_{\text{Ni}} \). This decrease can be attributed to a decrease in absorption when increasing the Ni thickness. To substantiate this claim, the absorption profile for back and front-pumping is calculated using a transfer matrix method,\(^{15,16}\) and the resulting profile for front-pumping is shown as an example in Fig. 3(a). Clearly, the amount of absorbed light per volume is decreased on increasing \( d_{\text{Ni}} \), caused by the increasing reflectivity of the film. On assuming that the maximum demagnetization scales linearly with the absorbed light, which experimentally has been verified for small laser fluences,\(^7\) it is expected that \( q_{\text{max}} \) in the front-pump and back-pump configuration is proportional to

\[
q_{\text{max,front}} \propto \frac{\int_0^{d_{\text{Ni}}} |E_{\text{front}}(x)|^2 |E_{\text{front}}(x)|^2 \ dx}{\int_0^{d_{\text{Ni}}} |E_{\text{front}}(x)|^2 \ dx},
\]

\[
q_{\text{max,back}} \propto \frac{\int_0^{d_{\text{Ni}}} |E_{\text{front}}(x)|^2 |E_{\text{back}}(x)|^2 \ dx}{\int_0^{d_{\text{Ni}}} |E_{\text{front}}(x)|^2 \ dx},
\]

where

\[
d_{\text{Ni}} = \frac{1}{2} \left( d_{\text{Ni}} + d_{\text{Ni}}' \right)
\]

for the front-pump and back-pump configurations, respectively.
where \( E_{\text{front}}(x) \) and \( E_{\text{back}}(x) \) are the electric fields in the Ni film for front-pumping and back-pumping, respectively. Fig. 3(b) displays experimental and calculated values for the normalized maximum quenching \( q_{\text{norm}} \). The measurements are normalized at \( d_{\text{Al}} = 6 \text{ nm} \), as the exact fluences for front- and back-pumping can vary due to, for example, differences in focus or the influence of the substrate. The experimental values of \( q_{\text{norm}} \) correspond reasonably well to the transfer matrix calculations, showing that the thickness dependence of \( q_{\text{max}} \) for both front- and back-pumping can be fully attributed to a difference in absorption.

Concluding, the measurements on the Ni film on an insulating substrate show no traces of any spin transport effects, hence the observed ultrafast demagnetization in ferromagnetic films on insulating substrates can only be interpreted by ultrafast local angular momentum transfer to another subsystem. That such an ultrafast transfer can be expected to occur on the timescales measured in the presented experiments has already been shown by various theoretical approaches.\(^5,7\)

Finally, note that our measurements do not exclude that ultrafast transport of heat by hot electrons plays an important role in the demagnetization process, which can alter the distribution of the locally deposited energy in the spin system.

In the foregoing measurements, spin transport can only induce a change in the magneto-optical signal due to an altered distribution of spins in the magnetic thin film, as the film is deposited on an insulating substrate. However, calculations by Battiato et al. show that the effect of superdiffusive spin transport should be significantly enhanced on adding a conducting buffer layer, which allows for transport of spins outside of the magnetic layer.\(^9\) To test this prediction, a 10 nm thick Ni film is grown on top of an Al wedge, where the Ni layer is deposited directly after the Al wedge, preventing oxidation of the Ni/Al interface. Front-pump measurements are performed for a varying thickness of the Al buffer layer \( d_{\text{Al}} \). A schematic overview of the performed experiment is depicted in the inset of Fig. 4(a).

Fig. 4(a) shows the measured TRMOKE traces for varying \( d_{\text{Al}} \). Measurements are only shown for Al thicknesses up to 7 nm, as for thicker layers the demagnetization signal became too small to detect. At first sight, the demagnetization traces are very similar to the ones in Figs. 2(a) and 2(b); a rapid demagnetization is followed by a more slow remagnetization. There is no apparent enhancement of the demagnetization due to the addition of the conducting buffer layer. This observation is supported by the data in Fig. 4(b), where \( \tau_m \) is plotted as a function of \( d_{\text{Al}} \). \( \tau_m \) does not change within the experimental accuracy on adding an Al bottom layer, and the demagnetization times are almost identical to the values obtained on the samples with only the Ni film on the insulating substrate.

The normalized maximum quenching, shown in Fig. 4(c), follows approximately the same trend as expected from the transfer matrix calculations. Again a thicker metallic layer increases the reflectivity and decreases the absorption per volume, and thus also the maximum quenching. A slight deviation in the experimental trend is visible for very small values of \( d_{\text{Al}} \), where up to 2 nm, an increase of the maximum quenching is observed. The origin of this slight increase could be speculated to be superdiffusive transport of majority charge carriers into the Al buffer layer. However, a more likely explanation is the altered growth of the Ni film on a thin Al film, yielding a smaller absorption or larger disorder, which results in slightly different magnetization dynamics. Nonetheless, it can be concluded that no significant enhancement of demagnetization due to superdiffusive spin transport is observed on adding the conducting buffer layer.

In conclusion, we studied the influence of superdiffusive transport on ultrafast laser-induced magnetization dynamics in Ni. By comparing front-pump with back-pump measurements, we concluded that transport plays no significant role in the demagnetization process of ferromagnetic thin films on insulating substrates. Furthermore, adding a conductive buffer layer did not show a significant enhancement of the demagnetization rate due to superdiffusive transport. This means that laser-induced demagnetization in the investigated films is dominated by an ultrafast local transfer of angular momentum away from the spin system. The measurements do not exclude the presence of superdiffusive spin transport in laser-induced magnetization dynamics, like in, for example, more complex stacks, but do show that it is not the dominant effect in simple ferromagnetic films.

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