Ultrafast demagnetization in iron: Separating effects by their nonlinearity

Kevin Bühlmann, Rafael Gort, Gerard Salvatella, Simon Däster, Andreas Fognini, Thomas Bähler, Christian Dornes, C. A. F. Vaz, Andreas Vaterlaus, and Yves Acremann

View Online   Export Citation   CrossMark

ARTICLES YOU MAY BE INTERESTED IN

Perspective: Ultrafast magnetism and THz spintronics
Journal of Applied Physics **120**, 140901 (2016); https://doi.org/10.1063/1.4958846

Ultrafast demagnetization by hot electrons: Diffusion or super-diffusion?
Structural Dynamics **3**, 055101 (2016); https://doi.org/10.1063/1.4964892

The influence of the excitation pulse length on ultrafast magnetization dynamics in nickel
Structural Dynamics **2**, 024501 (2015); https://doi.org/10.1063/1.4914891

Ultrafast demagnetization in iron: Separating effects by their nonlinearity

Kevin Bühlmann,[1,a)] Rafael Gort,[1] Gerard Salvatella,[1] Simon Däster,[1] Andreas Fognini,[1,2] Thomas Bähler,[1] Christian Dornes,[3] C. A. F. Vaz,[4] Andreas Vaterlaus,[1] and Yves Acremann[1]

[1]*Laboratory for Solid State Physics, ETH Zurich, 8093 Zurich, Switzerland*
[2]*Department of Quantum Nanoscience, TU Delft, 2600 GA Delft, The Netherlands*
[3]*Institute for Quantum Electronics, ETH Zurich, 8093 Zurich, Switzerland*
[4]*Swiss Light Source, Paul Scherrer Institut, PSI, 5232 Villigen, Switzerland*

The laser-driven ultrafast demagnetization effect is one of the long-standing problems in solid-state physics. The time scale is given not only by the transfer of energy, but also by the transport of angular momentum away from the spin system. Through a double-pulse experiment resembling two-dimensional spectroscopy, we separate the different pathways by their nonlinear properties. We find (a) that the loss of magnetization within 400 fs is not affected by the previous excitations (linear process), and (b) we observe a picosecond demagnetization contribution that is strongly affected by the previous excitations. Our experimental approach is useful not only for studying femtosecond spin dynamics, but can also be adapted to other problems in solid-state dynamics. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http:// creativecommons.org/licenses/by/4.0/). [https://doi.org/10.1063/1.5040344]

INTRODUCTION

In 1996, Beaurepaire *et al.*[1] discovered that demagnetization processes can occur within less than a picosecond, which is below the time responses typically associated with spin precession. In their paper, the three-temperature model (3TM) was introduced, where the couplings between the spin system, the electron gas, and the lattice are described in terms of energy transfer. Following their work, the mechanism responsible for femtosecond angular momentum transfer has been the focus of ultrafast spin dynamics research.

The laser-induced ultrafast loss of spin angular momentum can be attributed to two effects. (a) The electron spins can flip in the excited ferromagnet due to collisions with phonons[2] and hot electrons[3–5] by the Elliot-Yafet scattering mechanism.[6] Spin flip scattering in the bulk is fundamentally caused by spin-orbit coupling.[7–14] In addition, the temperature-dependent shifts of the chemical potentials for minority and majority electrons have been identified as a driving force for spin flips.[15] (b) Spin transport, whereby spin-polarized electrons are transported from the magnetic surface deeper into the sample, where they can flip their spin outside the region being probed.[16–18] This mechanism has been observed experimentally.[19–23] The spin current can even be injected into a second ferromagnet, where it affects the magnetization through the spin torque effect.[22–26] However, the spin current alone cannot fully explain ultrafast demagnetization, as shown by Wieczorek *et al.*[27]

Here, we present a dual pump-probe experiment that aims to investigate the nonlinear aspects of the ultrafast demagnetization effect. We use a first pump pulse P_h to heat the ferromagnet. This pulse arrives at time τ before time zero. Its energy is primarily absorbed by the electron gas, leading to an increase in its temperature. Within the electron-lattice equilibration time $\tau_{el} \approx 1.2$ ps (determined from the reflectivity signal according to Refs. 28 and 29), the

---

a)Electronic mail: kevinbu@phys.ethz.ch

electron gas equilibrates with the lattice to a common temperature. A second pump pulse P_d excites the sample at time zero. We observe the demagnetization caused by the second pump pulse. Here, we study how the previous excitations from the first pump pulse affects the demagnetization caused by the second pump pulse.

EXPERIMENTAL SETUP

The sample consists of a single-crystalline Fe layer grown on a substrate of MgO (001) by molecular beam epitaxy. The Fe layer is 17 nm thick, and it is capped by 2 nm MgO and 2 nm Al. The sample is placed inside a cryostat, which allows for cooling down to 10 K, to suppress excitations in the phononic, electronic, and spin system. The average temperature increases to 100 K once the pump laser beams are present. A static magnetic field of 350 Oe is applied, which saturates the magnetization along the easy axis, indicated by ↑, ↓. An amplified Ti:sapphire laser system with a repetition rate of 10 kHz and a pulse length of 25 fs FWHM is used to excite and detect the magnetization. The pump laser is split into heating and demagnetizing pulses, which are delayed independently. The pump section of the experiment was designed to provide equal dispersion for both pulses. In addition, separate compressors are used for the pump and probe pulses, which are optimized for the shortest pulses on the sample. The probe beam is converted from 800 nm to 400 nm using a beta barium borate (BBO) crystal to avoid state blocking effects.[30,31] We use the longitudinal magneto-optical Kerr rotation to detect the magnetization. The pump beam is modulated by a mechanical chopper at 83 Hz for lock-in detection of the pump-induced demagnetization. The signal is measured for the two magnetization directions ↑ and ↓, and the difference is calculated. This difference represents the laser-induced demagnetization of the sample ΔM.

EXPERIMENTAL RESULTS AND DISCUSSION

The demagnetization curves for different pump-pump delays τ are shown in Fig. 1. The fluences of both pump pulses are adjusted such that each of them alone demagnetizes the sample by 17%. This is apparent at $\tau = 50$ ps. Here, the heating pulse P_h causes approximately the same demagnetization as P_d. The time of 50 ps is sufficient to cause almost a complete
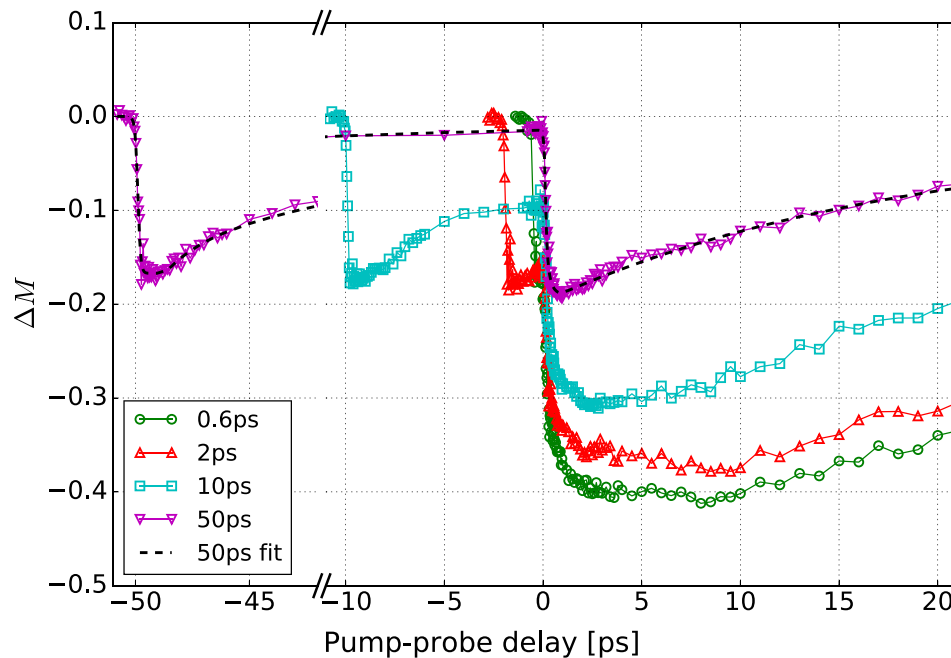

FIG. 1. Measured demagnetization ΔM caused by a pair of pump pulses (heating pulse P_h followed by the demagnetization pulse P_d). The excitations caused by P_h enhance the demagnetization of P_d. The legend indicates the time interval between the heating and demagnetization pulses, τ. The line for $\tau = 50$ ps shows the fit ΔM_{fit} used to calculate ΔM_d shown in Fig. 2.

recovery of the magnetization and significant cooling of the electron gas and lattice. However, if τ is reduced, then the heating pulse starts to enhance the demagnetization caused by P_d. In addition, for $\tau < 2$ ps, the largest demagnetization is not reached after the ultrafast drop near $t = 0$ but approximately 10 ps later.

To study the temporal behavior of ΔM, we subtract the background of the magnetization recovery caused by the heating pulse P_h. The demagnetization of a single pulse is fitted by the empirically determined function

$$\Delta M_{\text{fit}} = \frac{a_0}{2}\left(1 + \tanh\left(\frac{t - t_0}{\tau_0}\right)\right) + \Theta(t - t_1)a_1\left(e^{\frac{-(t-t_1)}{\tau_1}} - 1\right) + \Theta(t - t_2)a_2\left(e^{\frac{-(t-t_2)}{\tau_2}} - 1\right). \quad (1)$$

Here, $\Theta(x)$ is the Heaviside function. The first term fits the fast decay, and the second and third terms fit the recovery. This single-pulse fit is determined using the first part of the trace at $\tau = 50$ ps (before the second pulse hits) and is displayed in Fig. 1. ΔM_{fit} is shifted in time and subtracted from each measured demagnetization curve ΔM

$$\Delta M_d = \Delta M - \Delta M_{\text{fit}}. \quad (2)$$

The resulting function ΔM_d would be equal to ΔM caused by P_d alone if the magnetization reacted in a linear manner to the pump pulses. For small demagnetization amplitudes, this is actually the case, as demonstrated in Ref. 32. In contrast, we work with larger demagnetization amplitudes of 17% per pulse, driving the system into a nonlinear response regime.

The result is shown in Fig. 2. We distinguish between the initial, ultrafast part of the demagnetization at $t < 400$ fs and the dynamics occurring on a longer time scale of up to 10 ps. We do not observe a significant effect of the heating pulse P_h on ΔM_d for the ultrafast part of the demagnetization (visible in the inset of Fig. 2). All the measurements of ΔM_d for $t < 400$ fs are equal within the margin of error and are independent of the pump-pump delay time τ. The ultrafast demagnetization process is therefore linear within the accuracy of our experiment. This linear effect is not predicted by the magnetic three-temperature model.[2]
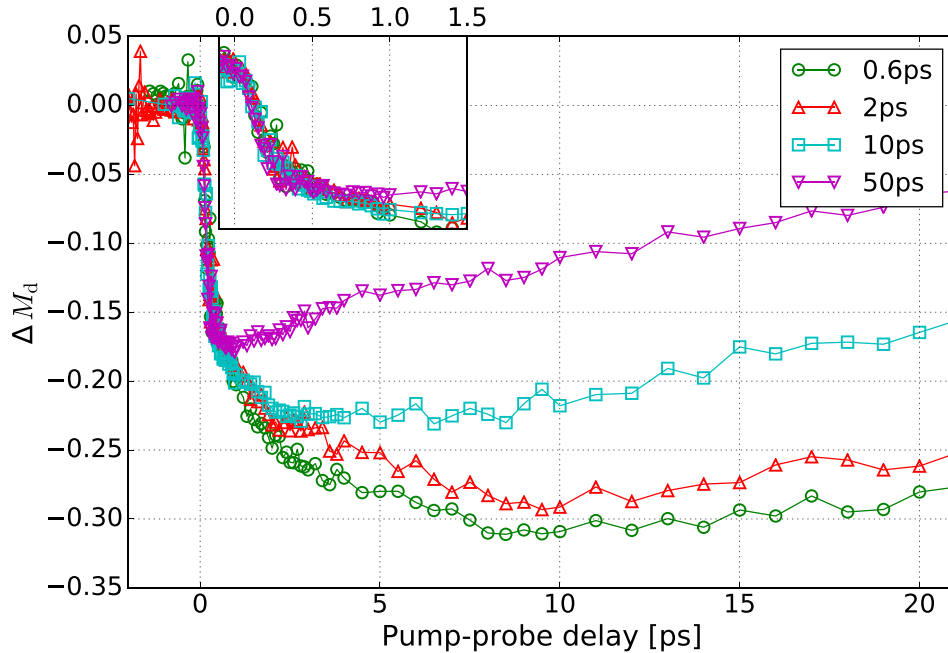

FIG. 2. Demagnetization ΔM_d caused by P_d after subtracting the demagnetization caused by the heating pulse P_h. The inset shows that the ultrafast part of the demagnetization is not affected by the heating pulse. However, on the picosecond time scale, we observe an enhancement in the demagnetization caused by P_h. In the case of a pump-pump delay of $\tau = 0.6$ ps, the maximum amplitude of ΔM_d is reached at $t \approx 10$ ps.

In contrast, the response for $t > 1$ ps strongly depends on τ and is therefore affected by the heating pulse, as shown in Fig. 2. The demagnetization is enhanced by the heating pulse P_h, and it reaches its maximum up to 10 ps after the demagnetizing pulse at $t = 0$ (for $\tau < 2$ ps).

The following question thus arises: which of the reservoirs (spin system, electron gas, or the lattice) excited by P_h causes the enhancement of the demagnetization? Here, we define the *enhancement* ΔM_e as the maximum deviation between the demagnetization with and without the heating pulse P_h

$$\Delta M_e(\tau) = -\max_t \left(|\Delta M_d(\tau, t)| - |\Delta M_{\text{no heating}}(t)| \right). \tag{3}$$

In order to keep the average heat load on the sample as well as the average temperature constant, the demagnetization without heating pulse has been determined by shifting the heating pulse to a time after the measurement pulse. The enhancement ΔM_e is plotted as a function of τ in Fig. 3. The strongest enhancement is observed for $\tau = 0.6$ ps at $t = 7$ ps.

We find a correlation between the enhancement ΔM_e and the demagnetization just before P_d: Fig. 3 shows a linear relation between the two quantities. This result suggests that magnetic excitations may be the source of the enhancement ΔM_e. This is consistent with the model proposed by Mueller *et al.*,[15] where they identify the magnetization-dependent shift of the exchange splitting as a feedback mechanism causing a larger separation of the spin-split chemical potentials and therefore more spin flips.

Cheng *et al.*[33] performed a similar double-pump - probe experiment on TbFeCo to investigate the nonlinear effects of the demagnetization process. Our results agree with their atomistic Landau-Lifshitz-Gilbert model, which shows that excitations of the spin system can lead to further demagnetization.

CONCLUSIONS

We conclude from our results that the ultrafast loss of the magnetization within the first 400 fs is not enhanced by the heating pulse. Thus, it is not affected by the previous generation
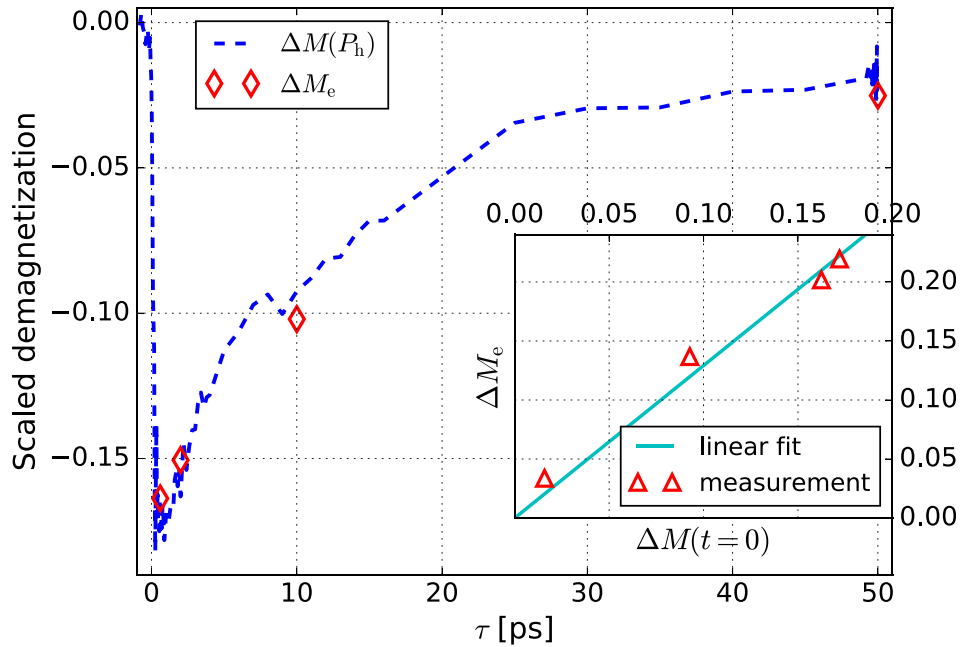

FIG. 3. Correlation between the enhancement $\Delta M_e(\tau)$ (diamonds) and the demagnetization caused by a single pump (dashed line) $\Delta M(P_h)$. Note that ΔM_e has been scaled in amplitude to match $\Delta M(P_h)$. The inset shows the linear relation between the measured demagnetization just before P_d is applied [given as $\Delta M(t = 0, \tau)$] and ΔM_e.

of hot electrons, phonons, or magnons within the margin of error of our experiment. A possible mechanism relevant on this time scale is the spin transport effect. However, we observe a significant (nonlinear) enhancement in the demagnetization on a longer time scale of up to 10 ps. The enhancement is proportional to the demagnetization caused by the first pump pulse, which indicates that the presence of disorder in the spin system enhances the spin flip probability.

Our results indicate that a new framework is needed to fully understand the observed ultrafast demagnetization phenomena, including the fully linear ultrafast contribution. The experimental results indicate that the mechanism for the ultrafast loss of the magnetization is different from the mechanism causing the slow drop on the picosecond time scale.[29] We suggest that novel spin- and time-resolved photoemission experiments will be able to separate and identify the two effects[34,35] and shed further light on the fundamental processes underlying the mechanism for ultrafast demagnetization.

Furthermore, our experiment shows that the nonlinear aspects of ultrafast processes in solids can be used to separate similar ultrafast contributions. Such an approach could be useful for other problems in condensed matter dynamics, particularly in the case of correlated systems.

ACKNOWLEDGMENTS

[1] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, "Ultrafast spin dynamics in ferromagnetic nickel," Phys. Rev. Lett. **76**, 4250–4253 (1996).

[2] B. Koopmans, G. Malinowski, F. Dalla Longa, D. Steiauf, M. Fähnle, T. Roth, M. Cinchetti, and M. Aeschlimann, "Explaining the paradoxical diversity of ultrafast laser-induced demagnetization," Nat. Mater. **9**, 259–265 (2010).

[3] B. Y. Mueller, T. Roth, M. Cinchetti, M. Aeschlimann, and B. Rethfeld, "Driving force of ultrafast magnetization dynamics," New J. Phys. **13**, 123010 (2011).

[4] A. B. Schmidt, M. Pickel, M. Donath, P. Buczek, A. Ernst, V. P. Zhukov, P. M. Echenique, L. M. Sandratskii, E. V. Chulkov, and M. Weinelt, "Ultrafast magnon generation in an fe film on cu(100)," Phys. Rev. Lett. **105**, 197401 (2010).

[5] M. Krauss, T. Roth, S. Alebrand, D. Steil, M. Cinchetti, M. Aeschlimann, and H. C. Schneider, "Ultrafast demagnetization of ferromagnetic transition metals: The role of the coulomb interaction," Phys. Rev. B **80**, 180407 (2009).

[6] D. Steiauf and M. Fähnle, "Elliott-yafet mechanism and the discussion of femtosecond magnetization dynamics," Phys. Rev. B **79**, 140401 (2009).

[7] K. Carva, M. Battiato, and P. M. Oppeneer, "*Ab Initio* investigation of the elliott-yafet electron-phonon mechanism in laser-induced ultrafast demagnetization," Phys. Rev. Lett. **107**, 207201 (2011).

[8] G. P. Zhang, W. Hübner, G. Lefkidis, Y. Bai, and T. F. George, "Reply: Is the controversy over femtosecond magneto-optics really solved?," Nat. Phys. **7**, 665–666 (2011).

[9] C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupulescu, E. F. Aziz, M. Wietstruk, H. A. Dürr, and W. Eberhardt, "Femtosecond modification of electron localization and transfer of angular momentum in nickel," Nat. Mater. **6**, 740–743 (2007).

[10] G. P. Zhang and W. Hübner, "Laser-induced ultrafast demagnetization in ferromagnetic metals," Phys. Rev. Lett. **85**, 3025–3028 (2000).

[11] J. Fabian and S. Das Sarma, "Spin relaxation of conduction electrons in polyvalent metals: Theory and a realistic calculation," Phys. Rev. Lett. **81**, 5624–5627 (1998).

[12] D. Steiauf, C. Illg, and M. Fähnle, "Extension of yafet's theory of spin relaxation to ferromagnets," J. Magn. Magn. Mater. **322**, L5 (2010).

[13] W. Töws and G. M. Pastor, "Many-body theory of ultrafast demagnetization and angular momentum transfer in ferromagnetic transition metals," Phys. Rev. Lett. **115**, 217204 (2015).

[14] T. Roth, A. J. Schellekens, S. Alebrand, O. Schmitt, D. Steil, B. Koopmans, M. Cinchetti, and M. Aeschlimann, "Temperature dependence of laser-induced demagnetization in ni: A key for identifying the underlying mechanism," Phys. Rev. X **2**, 021006 (2012).

[15] B. Y. Mueller, A. Baral, S. Vollmar, M. Cinchetti, M. Aeschlimann, H. C. Schneider, and B. Rethfeld, "Feedback effect during ultrafast demagnetization dynamics in ferromagnets," Phys. Rev. Lett. **111**, 167204 (2013).

[16] M. Battiato, K. Carva, and P. M. Oppeneer, "Superdiffusive spin transport as a mechanism of ultrafast demagnetization," Phys. Rev. Lett. **105**, 027203 (2010).

[17] A. J. Schellekens, W. Verhoeven, T. N. Vader, and B. Koopmans, "Investigating the contribution of superdiffusive transport to ultrafast demagnetization of ferromagnetic thin films," Appl. Phys. Lett. **102**, 252408 (2013).

[18] A. Fognini, T. U. Michlmayr, A. Vaterlaus, and Y. Acremann, "Laser-induced ultrafast spin current pulses: A thermodynamic approach," J. Phys.: Condens. Matter **29**, 214002 (2017).

[19] G. Malinowski, F. Dalla Longa, J. H. H. Rietjens, P. V. Paluskar, R. Huijink, H. J. M. Swagten, and B. Koopmans, "Control of speed and efficiency of ultrafast demagnetization by direct transfer of spin angular momentum," Nat. Phys. **4**, 855–858 (2008).

[20] A. Eschenlohr, M. Battiato, P. Maldonado, N. Pontius, T. Kachel, K. Holldack, R. Mitzner, A. Föhlisch, P. M. Oppeneer, and C. Stamm, "Ultrafast spin transport as key to femtosecond demagnetization," Nat. Mater. **12**, 332–336 (2013).

[21]T. Kampfrath, M. Battiato, P. Maldonado, G. Eilers, J. Notzold, S. Mahrlein, V. Zbarsky, F. Freimuth, Y. Mokrousov, S. Blugel, M. Wolf, I. Radu, P. Oppeneer, and M. Munzenberg, "Terahertz spin current pulses controlled by magnetic heterostructures," Nat. Nanotechnol. **8**, 256–260 (2013).

[22]D. Rudolf, C. La-o-vorakiat, M. Battiato, R. Adam, J. M. Shaw, E. Turgut, P. Maldonado, S. Mathias, P. Grychtol, H. T. Nembach, T. J. Silva, M. Aeschlimann, H. C. Kapteyn, M. M. Murnane, C. M. Schneider, and P. M. Oppeneer, "Ultrafast magnetization enhancement in metallic multilayers driven by superdiffusive spin current," Nat. Commun. **3**, 1037 (2012).

[23]E. Turgut, C. La-o-vorakiat, J. M. Shaw, P. Grychtol, H. T. Nembach, D. Rudolf, R. Adam, M. Aeschlimann, C. M. Schneider, T. J. Silva, M. M. Murnane, H. C. Kapteyn, and S. Mathias, "Controlling the competition between optically induced ultrafast spin-flip scattering and spin transport in magnetic multilayers," Phys. Rev. Lett. **110**, 197201 (2013).

[24]G.-M. Choi, B.-C. Min, K.-J. Lee, and D. G. Cahill, "Spin current generated by thermally driven ultrafast demagnetization," Nat. Commun. **5**, 4334 (2014).

[25]A. J. Schellekens, K. C. Kuiper, R. de Wit, and B. Koopmans, "Ultrafast spin-transfer torque driven by femtosecond pulsed-laser excitation," Nat. Commun. **5**, 4333 (2014).

[26]J. Kimling and D. G. Cahill, "Spin diffusion induced by pulsed-laser heating and the role of spin heat accumulation," Phys. Rev. B **95**, 014402 (2017).

[27]J. Wieczorek, A. Eschenlohr, B. Weidtmann, M. Rösner, N. Bergeard, A. Tarasevitch, T. O. Wehling, and U. Bovensiepen, "Separation of ultrafast spin currents and spin-flip scattering in co/cu(001) driven by femtosecond laser excitation employing the complex magneto-optical kerr effect," Phys. Rev. B **92**, 174410 (2015).

[28]R. W. Schoenlein, W. Z. Lin, J. G. Fujimoto, and G. L. Eesley, "Femtosecond studies of nonequilibrium electronic processes in metals," Phys. Rev. Lett. **58**, 1680–1683 (1987).

[29]E. Carpene, E. Mancini, C. Dallera, M. Brenna, E. Puppin, and S. De Silvestri, "Dynamics of electron-magnon interaction and ultrafast demagnetization in thin iron films," Phys. Rev. B **78**, 174422 (2008).

[30]B. Koopmans, M. van Kampen, J. T. Kohlhepp, and W. J. M. de Jonge, "Ultrafast magneto-optics in nickel: Magnetism or optics?," Phys. Rev. Lett. **85**, 844–847 (2000).

[31]P. M. Oppeneer and A. Liebsch, "Ultrafast demagnetization in ni: Theory of magneto-optics for non-equilibrium electron distributions," J. Phys.: Condens. Matter **16**, 5519 (2004).

[32]A. Fognini, G. Salvatella, R. Gort, T. Michlmayr, A. Vaterlaus, and Y. Acremann, "The influence of the excitation pulse length on ultrafast magnetization dynamics in nickel," Struct. Dyn. **2**, 024501 (2015).

[33]T. Cheng, J. Wu, T. Liu, X. Zou, J. Cai, R. W. Chantrell, and Y. Xu, "Dual-pump manipulation of ultrafast demagnetization in TbFeCo," Phys. Rev. B **93**, 064401 (2016).

[34]M. Plötzing, R. Adam, C. Weier, L. Plucinski, S. Eich, S. Emmerich, M. Rollinger, M. Aeschlimann, S. Mathias, and C. M. Schneider, "Spin-resolved photoelectron spectroscopy using femtosecond extreme ultraviolet light pulses from high-order harmonic generation," Rev. Sci. Instrum. **87**, 043903 (2016).

[35]S. Eich, M. Plötzing, M. Rollinger, S. Emmerich, R. Adam, C. Chen, H. C. Kapteyn, M. M. Murnane, L. Plucinski, D. Steil, B. Stadtmüller, M. Cinchetti, M. Aeschlimann, C. M. Schneider, and S. Mathias, "Band structure evolution during the ultrafast ferromagnetic-paramagnetic phase transition in cobalt," Sci. Adv. **3**, e1602094 (2017).