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Understanding the loss of magnetic order and the microscopic mechanisms involved in laser induced magnetization dynamics is one of the most challenging topics in today's magnetism research. While scattering between spins, phonons, magnons and electrons have been proposed as sources for dissipation of spin angular momentum, ultrafast spin dependent transport of hot electrons has been pointed out as a potential candidate to explain ultrafast demagnetization without resorting to any spin dissipation channel. Here we use time resolved magneto-optical Kerr measurements to extract the influence of spin dependent transport on the demagnetization dynamics taking place in magnetic samples with alternating domains with opposite magnetization directions. We unambiguously show that whatever the sample magnetic configuration, the demagnetization takes place during the same time, demonstrating that hot electrons spin dependent transfer between neighboring domains does not alter the ultrafast magnetization dynamics in our systems with perpendicular anisotropy and 140 nm domain sizes.

Since the first experimental proof of ultrafast demagnetization in 1996 by Beaurepaire *et al.*[1], laser induced magnetization dynamics has been the subject of abundant theoretical and experimental studies. The motivation is driven by the twofold challenge of understanding magnetization dynamics in a strongly out-of-equilibrium system and optically controlling the magnetic state of a nanostructure on the shortest timescale. Apart from the potential applications in magnetic storage media and other spintronic devices, the understanding of this phenomenon remains incomplete and no consensus on the importance of the different microscopic mechanisms involved in the ultrafast magnetization dynamics has been achieved so far.

Indeed, different mechanisms have been proposed to explain the loss of magnetization following a strong laser pulse excitation. Initially, they were based on the conservation of angular momentum during the demagnetization process. Some of them relied on the coupling between the photon field and the spin bath[2,3] while others were founded on spin-flip processes occurring when electrons scatter with electrons[4], phonons[5–13] or magnons[14,15]. Recently, the role of optically induced ultrafast spin transport on the magnetization dynamics started to be addressed. It was first shown by Malinowski *et al.*[16] that hot electrons spin transport enhances and speeds up the demagnetization in a spin valve structure as compared to a single magnetic layer. Thereafter, Battiato *et al.* developed a semi-classical model based on optically excited spin polarized electron transport in the superdiffusive regime[17]. The spin-dependent lifetimes and velocity of optically excited hot electrons result in spin currents within a ferromagnetic layer, inducing a modification of the magnetization profile within the layer on the femtosecond timescale. Consequently, it was argued that superdiffusive spin transport may alone be responsible for the ultrafast magnetization dynamics[18]. The existence of laser induced hot electron spin transport was then clearly observed by Melnikov *et al*[19].

The recent advent of techniques combining femtosecond time resolution and element-specific measurement of the magnetization such as synchrotron radiation using femtoslicing[7,8], free-electron lasers[20,21] and high-harmonic generation[22,23] sources has allowed tremendous progress in this field of research. For instance, Mathias *et al.* have

shown that two entities in a strongly coupled ferromagnetic alloy can have their own demagnetization dynamics[24]. In the case of ferrimagnetic alloys, the different dynamics of both sublattices has been shown to be an important ingredient in laser induced magnetization reversal[25]. Regarding optically excited hot electrons, it was shown that hot electron spin transport on the femtosecond timescale could not only enhance the demagnetization but could also increase the magnetization of a buried layer in a magnetic multilayer depending on the relative magnetization orientation in the different layers[26]. By controlling the inter-layer spin current, it was demonstrated that both spin transport and spin-flip scattering processes contribute to the ultrafast magnetization dynamics[27]. However, in the case of single magnetic layers, the importance of ultrafast spin transport remains unclear[28]. While all these works dealt with spin transport taking place along the multilayers thickness, an interesting experiment was carried out by Pfau *et al.*[29] and Vodungbo *et al.*[30] who studied the influence of superdiffusive transport on the demagnetization in the presence of nanoscale magnetic domains. In this case, the samples are chemically homogeneous and the boundaries between magnetic domains are formed by magnetic domain walls (DW). It was shown that hot electrons spin transport induced a modification of the magnetic form factor due to spin accumulation around the DW, resulting in a change of the magnetic scattering pattern[29]. By studying the evolution of the demagnetization time (τ_M) as a function of the magnetization quenching, Vodungbo *et al.* concluded that superdiffusive spin currents were responsible for the constant demagnetization time independently of the laser fluence[30].

In this paper, we report on the influence of the domain configuration in ferromagnetic structures on the ultrafast magnetization dynamics. We show that the demagnetization time increases with the laser fluence when the sample magnetization is saturated, similarly to what is usually observed in transition metals. Furthermore, our results reveal that the demagnetization time does not evolve when the sample magnetic configuration goes from saturation to a multidomain state, questioning the role of hot electrons spin transport on the ultrafast demagnetization in such a system.

Results

Multilayer fabrication and static characterization. The samples used in this study consist of a two different multilayers $[(\text{Co}\ (0.4\ \text{nm})/\text{Pd}\ (0.6\ \text{nm})]_{\times 20}$ (CoPd multilayer) and $[(\text{Co}\ (0.5\ \text{nm})/\text{Pt}\ (1.0\ \text{nm})]_{\times 15}$ (CoPt multilayer) grown at room temperature by dc magnetron sputtering. The static magnetic properties were analyzed by magneto-optical Kerr effect and magnetic force microscopy. Figure 1b and 1c show hysteresis loops measured on both samples with a magnetic field applied perpendicularly to the sample plane. From these measurements, it is clear that the CoPd and CoPt multilayers exhibit an out-of-plane anisotropy with a domain maze structure at low applied magnetic field showing alternating up and down magnetization (Fig. 1a). Indeed, when the field is reduced from saturation, a large jump in the Kerr signal is observed even though the magnetic field is not reversed. This jump is due to the onset of the domain formation. The onset field values are slightly different for the CoPd multilayers (80 mT) and the CoPt multilayers (50 mT) due to their different saturation magnetization and effective anisotropy. Magnetic force microscopy images obtained for both samples are depicted in the insets of figure 1b and 1c. Both samples show similar domain configurations with domain sizes of 145 nm and 132 nm for the CoPt and CoPd multilayers, respectively. Furthermore, by modifying the amplitude of the applied magnetic field, it is possible to control the magnetic structure of our samples, i.e. the ratio between the size of domains with opposite magnetization direction.

Magnetic domain structure and ultrafast demagnetization. To investigate the role of hot electron spin transport between neighboring domains, we performed a systematic study of the demagnetization occurring in our multilayers after a laser pulse excitation for a large panel of fluences and applied magnetic fields. The magnetization dynamics was studied by polar time resolved magneto-optical Kerr effect (TRMOKE) measurements. The samples were pumped by an infrared laser pulse at 800 nm wavelength with a pulse duration of 50 ± 5 fs. The probe beam was frequency doubled (400 nm wavelength) in order to minimize optical artifacts such as dichroic bleaching[31]. In this configuration, the measured Kerr

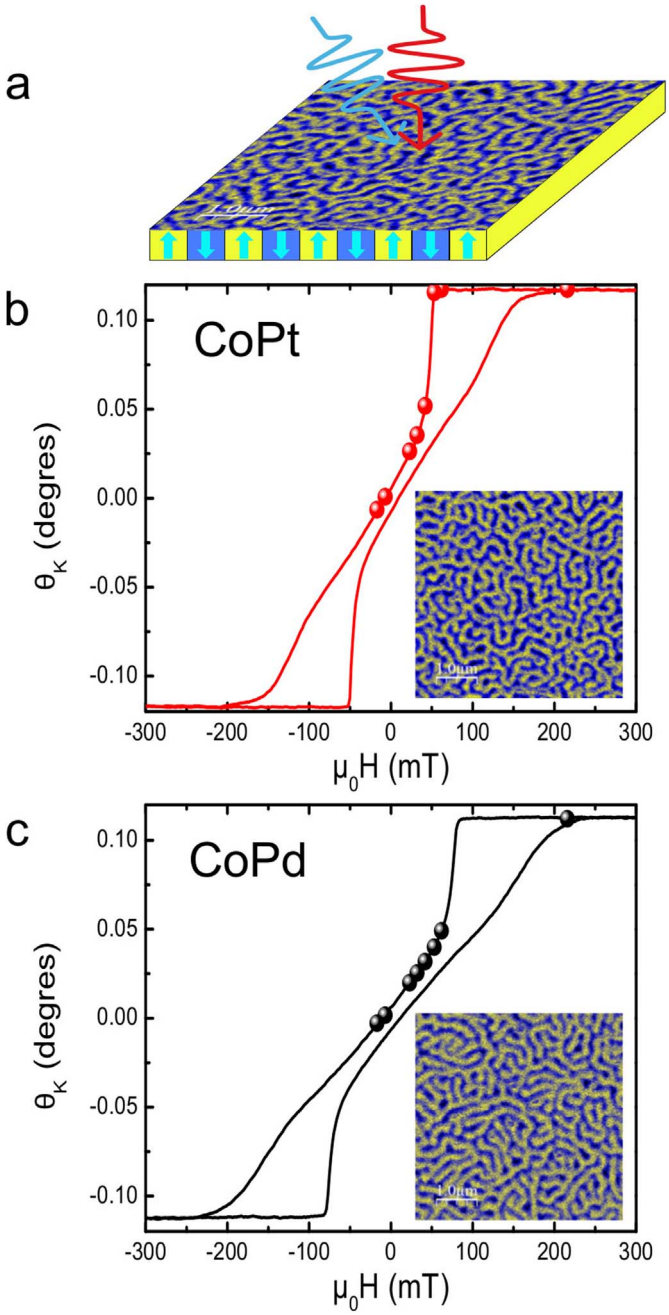

Figure 1 | (a) Schematic representation of the sample magnetic domain structure. Blue and yellow areas represent magnetic domains with opposite magnetization direction perpendicular to the film plane. The blue and red curved arrows schematically show the probe and pump laser pulses, respectively. Magnetic hysteresis loop showing the variation of the Kerr rotation θ_K with the applied magnetic field for the (b) CoPt and (c) CoPd multilayers. The filled circles represent values of the Kerr rotation at a given field for which time resolved measurements were performed.(insets) Magnetic force microscopy images measured at zero applied field on the (b) CoPt and (c) CoPd multilayers. The images size is 5 × 5 μm².

rotation is proportional to the out-of-plane component of the magnetization. The measurements were performed for a wide range of fluences (0.35–5.6 mJ/cm^2) and for different amplitudes of the magnetic field applied perpendicularly to the sample plane (0–300 mT).

To start with, we performed time resolved experiment as a function of the laser fluence under a strong applied magnetic field to saturate the samples magnetization. The results are presented in fig. 2. Both multilayers show similar demagnetization curves with a fast loss of the Kerr signal occurring within the first hundreds of femtoseconds and followed by a slow magnetization recovery after few picoseconds (Fig. 2a,b). The absorption of a strong laser pulse results in a quasi-instantaneous increase of the electronic temperature. The energy is rapidly transferred to the spin bath leading to the fast drop of the magnetic signal. Equilibration between the electrons and phonons baths due to the electron-phonon interaction leads to the thermalization of the spin bath and a partial recovery of the magnetization. It is interesting to note that the time needed to demagnetize the sample increases with the demagnetization amplitude and so, with the fluence. Fig. 2c reports the evolution of the demagnetization time obtained by fitting the data using the three temperatures model (see Methods) as a function of the demagnetization amplitude[1,32]. We can clearly observe a large increase of the demagnetization time with the quenching of the Kerr signal for both CoPd and CoPt multilayers. This trend is similar to what has been previously observed in transition metals thin films[9]. The existence of a difference in demagnetization time for both multilayers might come from the difference in relative thickness between the Co and the Pd or Pt layers inducing different spin flip probabilities.

In order to study the influence of the magnetic domains configuration on the ultrafast demagnetization in these multilayers, we performed TRMOKE measurements for different applied magnetic fields. The measurements were performed at a fixed laser fluence to avoid spurious variation of the demagnetization time. Therefore, it is possible to carefully control the magnetic domain structure while measuring the ultrafast demagnetization. Fig. 3a shows measurements obtained on the CoPd and CoPt multilayers and for different values of the applied magnetic field. While above 80 mT (50 mT) the CoPd (CoPt) multilayer is completely saturated, reducing the applied magnetic field leads to the appearence of a magnetic domain network configuration. Time resolved measurements performed for these different applied fields clearly look identical, showing no influence of the domain structure on the ultrafast demagnetization (Fig. 3a). To get a quantitative estimate of the role played by hot electrons spin transport in our systems, we extracted the demagnetization time from these measurements. The evolution of the demagnetization time as a function of the magnetic configuration is plotted in Fig. 3b for CoPd and CoPt multilayers, for a laser fluence of 4.9 mJ/cm^2. We used the largest fluence available to maximise the number of excited electrons and because a higher fluence results in a longer demagnetization time as shown in Figure 2c. The CoPt and CoPd demagnetization times, i.e. $\tau_M^{CoPt} = 200$fs and $\tau_M^{CoPd} = 180$fs remain constant independently of the magnetic domain structure, indicating no or small contribution of the hot electrons spin transport between neighboring domains.

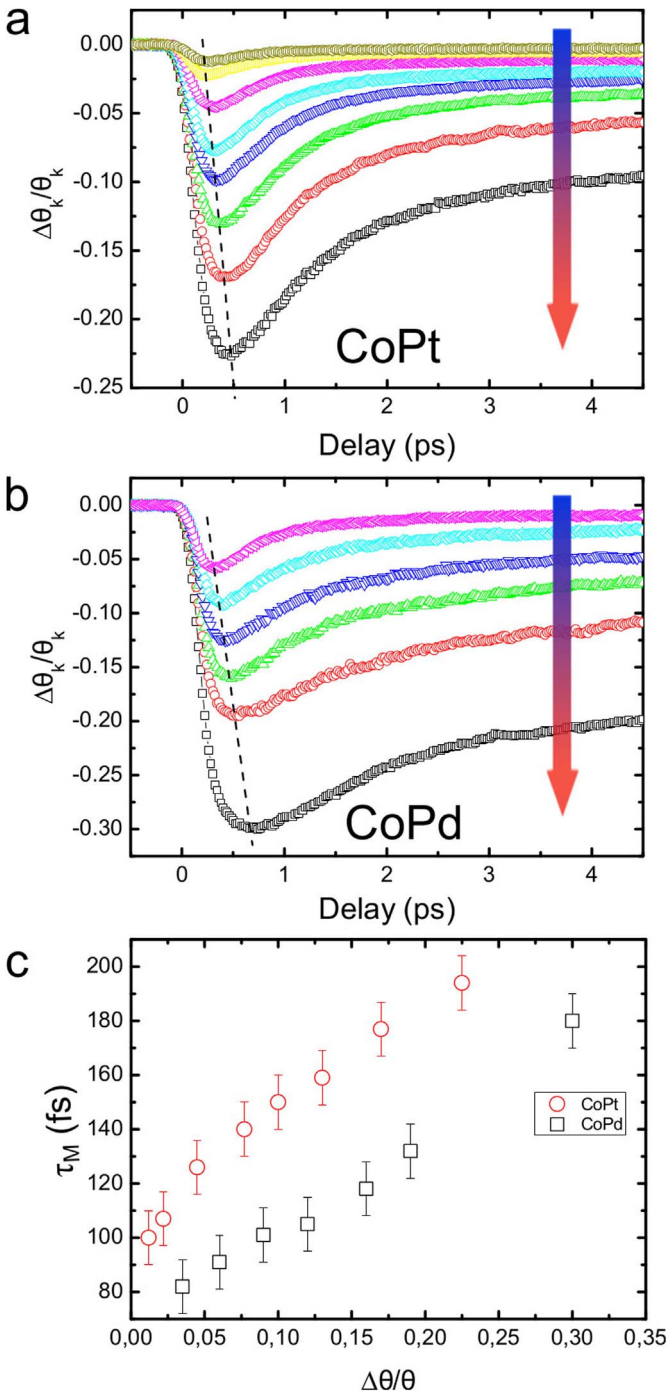

Figure 2 | Time resolved Kerr signal measured for different laser fluences on (a) the CoPt mulitlayers and (b) on the CoPd multilayers for various laser fluences (black 4.9 mJ/cm^2, red 4.2 mJ/cm^2, green 3.5 mJ/cm^2, blue 2.8 mJ/cm^2, cyan 2.1 mJ/cm^2, magenta 1.4 mJ/cm^2, yellow 0.7 mJ/cm^2, dark yellow 0.35 mJ/cm^2). The colored arrows schematically represent the increase in laser fluence with blue and red corresponding to low and high fluence, respectively. (c) Evolution of the demagnetization time as a function of the quenching of the Kerr signal for the CoPd (black squares) and CoPt (red circles) multilayers.

Discussion

To understand the role played by the hot electrons displacement between neighboring domains on the ultrafast demagnetization dynamics, we need to take into account that hot electrons transport properties are spin dependent. Indeed, in a magnetic material, spin-minority (minority) and spin-majority (majority) hot electrons possess different lifetimes and velocities[33,34]. Generally, minority hot electrons have a shorter lifetime and a lower velocity compared to majority hot electrons. This implies that most of the hot electrons crossing the DW will be majority hot electrons. However, once the DW is passed, majority hot electrons suddenly become minority hot electrons in the neighboring domain. Since they have much smaller lifetime and velocity, they will induce a minority spin accumulation in an area close to the DW, leading to a local reduction of the magnetization.

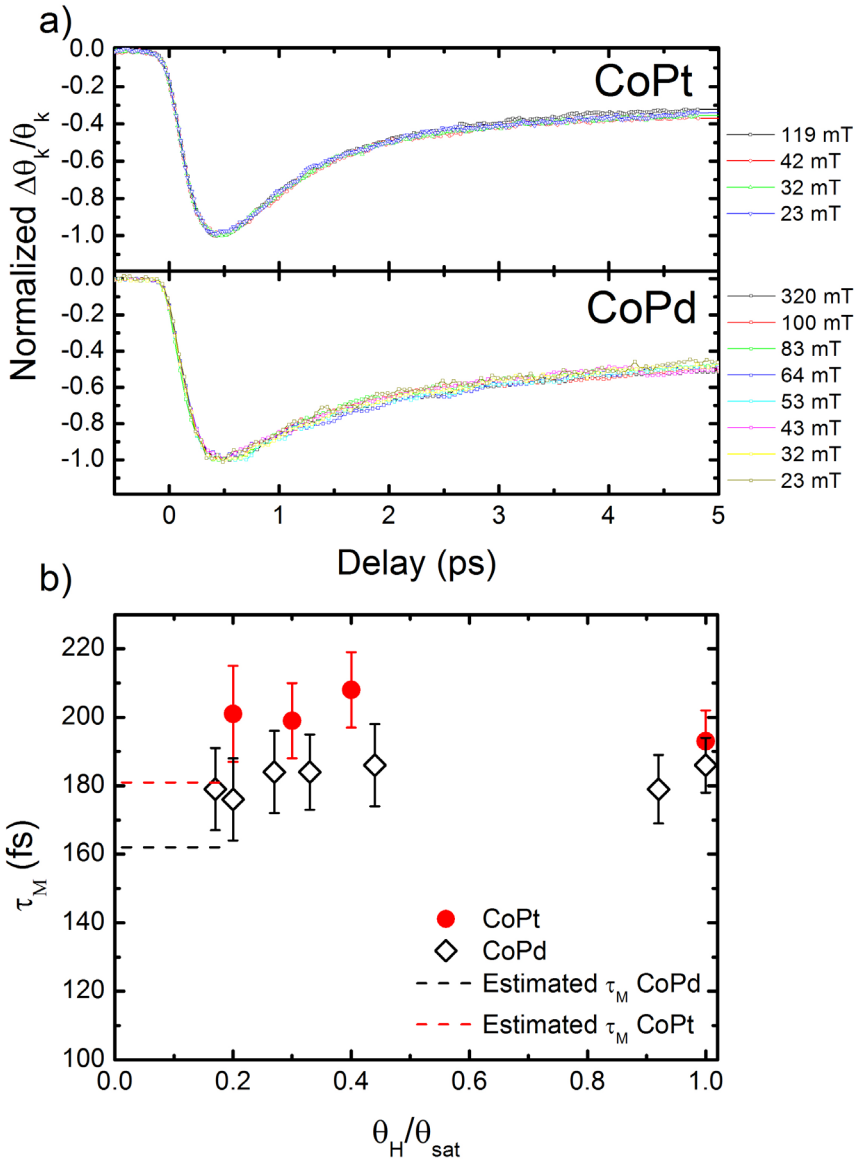

Figure 3 | (a) Time dependance of the Kerr signal measured on the CoPt and CoPd multilayers measured for different applied fields for a laser fluence of 4.9 mJ/cm^2. (b) Dependance of the demagnetization time as a function of the initial magnetic state represented by the ratio of the Kerr signal θ_H obtained for an applied field $\mu_0 H$ over the Kerr signal measured at saturation for the CoPt (red filled circles) and CoPd (open diamond) multilayers for a laser fluence of 4.9 mJ/cm^2. The red (black) dashed line corresponds to the estimated demagnetization time (see data analysis in the Methods section).

In their paper, Pfau and collaborators evaluated the spin accumulation at the domain wall boundary using the superdiffisuve model[17,29]. It was shown to result in a broadening of the magnetization profile around the domain wall center with a spatial extent of 20 nm resulting in the modification of the DW scattering form factor and thus leading to a shift of the maximum scattering intensity. Therefore, the demagnetization signal and a possible alteration of the demagnetization time due to the hot electron spin transport between neighboring domains should only come from δ = 20 nm around the DW.

The dynamics due to hot electron spin transport should alter the demagnetization curve shape and result in an increase of the demagnetization at the peak position. This extra demagnetization has been shown to vanish within the electron-phonon relaxation time (about 1 ps)[16]. However, no modification of the dynamics can be observed in the present study (Fig. 3a). Taking into account the noise level in our measurements, we can estimate the maximum proportion of the signal due to superdiffusive transport. The noise level can be estimated to be 3.5% and 5% for the CoPt and CoPd multilayers measurements, respectively. Using equation 4 (the supporting analysis to equation 4 can be found in the "Methods" section), the demagnetization signal due to superdiffusive spin currents are smaller than 33% and 25% of the total demagnetization signal within the 20 nm around the DW for CoPd and CoPt multilayers, respectively.

In the same way, we can estimate the contribution of hot electron spin transfer on the total demagnetization time. With the domain sizes in our CoPd and CoPt multilayers being 132 and 145 nm respectively, and assuming that hot electron spin transport result in a constant demagnetization time of τ_{sc} = 100 fs in both systems[30], the reduction of the effective demagnetization time between the saturated and the multidomain states would be 19 fs and 18 fs for CoPt and CoPd respectively, as represented by the dashed lines in Figure 3b (see data analysis in the Methods section for more details). In both cases, the expected decrease of the demagnetization time due to hot electrons transport is much larger than the error bars. We can thus conclude that the contribution of hot electrons spin transport on the demagnetization is negligible in our systems. As a result, the time resolved signal we measure by time resolved magneto-optical Kerr

effect is mainly reflecting demagnetization due to spin flip processes. Similar arguments can be used to analyze magnetic scattering experiments if only the amplitude of the signal is taken into account[30]. Even though, in the study of Vodungbo *et al.* the samples show a magnetic structure with domain sizes of 80 nm, using similar arguments as above, an increase of the demagnetization time with the laser fluence is still expected, contrary to what has been observed. Furthermore, our results clearly contrast with recent theoretical works supporting spin transport as the only relevant process in ultrafast demagnetization[17,18].

Our conclusions also contrast with experiments realized on magnetic spin-valve like multilayers for which a clear evidence of an ultrafast spin transport resulting in a modification of the magnetization dynamics was observed[16,19,26,27]. However, in that case, the different magnetic layers are usually only few nm thick comparable with the inelastic mean free path of hot electrons[33]. The spin accumulation due to hot electrons spin transport takes place over the full thickness of the layers inducing a demagnetization signal of the same order as the one generated by spin-flip scattering processes.

Finally, we would like to stress that even though many theoretical and experimental works have focused on hot electron spin lifetime in magnetic materials[33–36], the continuous variation of the local spin orientation in the DW induces a rotation of the exchange field which might significantly influence the spin lifetime. For instance, it was recently shown that the spin diffusion of conduction electrons by a magnetic DW strongly depends on the DW structure, and more precisely on the magnetization gradient present over distances comparable to characteristic lengths such as the spin diffusion length[37]. However, while they could be of prime importance in determining the spin accumulation length around the magnetic DW, the effects of a magnetization gradient on the diffusion of hot electron spins remain an unexplored area.

To conclude, we studied the demagnetization dynamics in CoPt and CoPd multilayers as a function of their magnetic configurations. When the samples are magnetically saturated, their demagnetization dynamics is similar to what is usually observed in 3d metals, namely, an increase of the demagnetization time is witnessed when the loss of magnetization increases. By measuring the demagnetization dynamics as a function of the domain structure, the extracted constant demagnetization time measured for different magnetic configurations reflects the absence of a modification of the demagnetization dynamics by hot electron spin transport in our samples. The analysis of the demagnetization curves shows that only a small part of the signal can be attributed to inter-domains hot electron spin transfer. Altogether, we showed that the role of inter-domains hot electrons spin transport on the demagnetization time is negligible. Reducing the domain size down to a length comparable to the spin accumulation length could potentially lead to a modification of the demagnetization dynamics due to inter-domain hot electron spin transport.

Methods

Sample preparation. The samples used in this study consist of two different multilayers SiN 30/Al 10/[Co 0.4/Pd 0.6]$_{\times 20}$/Pd 2 and SiOx/Ta 5/Pt 5/[Co 0.5/Pt 1.0]$_{\times 15}$/Pt 5 (thicknesses in nm) grown at room temperature by dc magnetron sputtering.

Experimental methods. The static polar Kerr loops presented in Fig. 1 were measured at a wavelength of 543 nm. The pump-probe measurements were carried out using a Femtolaser XL 300 pulsed laser with a wavelength of 800 nm and a pulse duration of about 50 ± 5 fs at a repetition rate of 5 MHz. The probe beam is frequency doubled (400 nm) in order to minimize optical artifacts such as dichroic bleaching[31]. The pump is much stronger than the probe with an intensity ratio of at least 20 for the lowest pump fluence. The pump laser is focused down to a spot size of about ~30 μm while the probe beam is focused down to about ~20 μm. Both beams are impinging on the sample almost at normal incidence with an angle of about ~5 degrees between them. In this configuration, the measured Kerr rotation is proportional to the out-of-plane component of the magnetization. The pump fluence is in the range of 0.5 to 4.9 mJ/cm^2.

Data analysis. The spin dynamics submitted to a laser pulse excitation can be described by a phenomenological thermodynamic model, i.e. the so-called three-temperatures model[1,32]. This model supposes that the system is split in three baths: the electron, lattice and spin baths. The laser photons are directly absorbed by the electronic system instantaneously creating hot electrons. Once thermalization is achieved by electron-electron interactions, each bath can be described by its own thermodynamic temperature. Then, the relaxation takes place by energy transfer between the different baths to balance their temperatures. The set of differential equations describing the heat flow between the three sub-systems are solved neglecting the spin specific heat, assuming that the measurements are done in the low laser fluence limit and that the electron temperature rises instantaneously upon laser excitation:

$$-\frac{\Delta M_z}{M_z} = [(\frac{A_1}{(t/\tau_0+1)^{1/2}} - \frac{(A_2\tau_E - A_1\tau_M)e^{-\frac{t}{\tau_M}}}{\tau_E-\tau_M} - \frac{\tau_E(A_1-A_2)e^{-\frac{t}{\tau_E}}}{\tau_E-\tau_M})\Theta(t)]\otimes\Gamma(t) \quad (1)$$

where $\Gamma(t)$ is the Gaussian laser pulse, $\Theta(t)$ is the step function and $\delta(t)$ is the Dirac delta function. The constant A_1 represents the value of $-\frac{\Delta M_z}{M_z}$ after equilibrium between electrons, spins and lattice is restored. The constant A_2 is proportional to the initial electron temperature rise. τ_M and τ_E are the demagnetization time and the relaxation time, respectively.

Assuming that the spin flip processes and hot electron spin transport contributions to the demagnetization time are independent we can calculate the effective demagnetization time as:

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_{sf}} + \frac{1}{\tau_{sc}} \quad (2)$$

where τ_{sf} and τ_{sc} are the demagnetization times due to spin-flip processes and hot electron spin transport, respectively. In such case and for $\tau_{sf}^{CoPt} = 200$ fs and $\tau_{sf}^{CoPd} = 180$ fs as measured in the saturated state (Fig. 3b) and $\tau_{sc} = 100$ *fs*[30], the total demagnetization time τ_M is given by:

$$\tau_M = \frac{(d-\delta) * \tau_{sf} + \delta * \tau_{eff}}{d} \quad (3)$$

with d being the domain size and $\delta = 20$ nm as the spin accumulation spatial extent around the domain wall. We can therefore estimate the demagnetization time for CoPt, $\tau_M^{CoPt} = 181$ fs, and CoPd, $\tau_M^{CoPd} = 162$ fs, multilayers in the multidomain state. This corresponds to a reduction of 19 and 18 fs for CoPt and CoPd compared with the demagnetization time measured in the saturated state.

Similarly, the total demagnetization due to both spin flip processes and spin dependent transport can be expressed as:

$$\Delta M = \frac{(d-\delta) * \delta M_{sf} + \delta * (\delta M_{sf} + \delta M_{sc})}{d} \quad (4)$$

where δM_{sf} and δM_{sc} are the demagnetization amplitudes due to spin flip processes and hot electrons spin depedent transport, respectively.

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Author contributions

G.M., N.M. and A.T. designed and coordinated the project. M.H., B.V. and J.L. made the samples. N.M., J.M. and G.M. performed the measurements. N.M. and G.M. analyzed the data and all the authors discussed the results. G.M. wrote the core of the manuscript while N.M., M.H., B.V., J.L., A.T., S.M. and E.E.F. contributed to certain parts of it.

Additional information

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