Hot-electrons-induced ultrafast demagnitization in Co/Pt multilayers

Hot electrons induced ultrafast demagnetization in Co/Pt multilayers

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Abstract

Using especially engineered structures to tailor the optical absorption in a metallic multilayer, we analyze the magnetization dynamics of a (Co/Pt) multilayer buried below a thick Cu layer. We demonstrate that hot electrons alone can very efficiently induce ultrafast demagnetization. Simulations based on hot electron ballistic transport implemented within a microscopic model that accounts for local dissipation of angular momentum nicely reproduce the experimental results, ruling out contribution of pure thermal transport.

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Ever since the discovery in 1996 by Beaurepaire *et al.* that magnetization in a magnetic material can be quenched within less than a picosecond using femtosecond laser pulses, the mechanisms responsible for the ultrafast loss of magnetization has been strongly debated [1]. During the last ten years, the focus has been put on the interplay between local dissipation of angular momentum and transport effects triggered by femtosecond laser pulses in magnetic multilayers.

Initially, laser induced demagnetization was explained in terms of heat transfer between electrons, phonons and spin baths within the so-called three temperatures model [1]. Lately, different models have been suggested relying either on a direct coupling between the photon field and the spin bath [2, 3], on spin-flip processes induced by electron scattering with particles or quasi-particules [4–14], or explaining the loss of demagnetization considering thermal mechanisms [15, 16].

During the last 25 years, transport and relaxation mechanisms of photogenerated carriers in metals have been extensively studied [17, 18] but the potential of laser-induced spin currents has only recently been signaled [19–26]. Malinowski *et al.* first pointed out that laser excited spin-polarized hot electron could increase and speed up the loss of magnetization in magnetic multilayers [19]. Thereafter, a model of ultrafast demagnetization involving superdiffusive transport of hot electron was reported by Battiato *et al.* [20, 21]. According to this model, energy and spin-dependent lifetime and velocity in a ferromagnetic layer could lead to the generation of a pure spin current after laser excitation resulting in the accumulation of minority spins in the ferromagnetic material when deposited on a spin sink, e.g. a metallic layer. It was even speculated whether superdiffusive hot electron spin transport could act as the only source of demagnetization without needing any local angular momentum relaxation channel. Following those ideas, many different groups reported on laser induced hot electron spin transport [22, 23]and subsequent spin transfer torque [24] as well as spin transfer torque due to laser induced thermal gradients [25, 26].

In all these experiments, demagnetization was induced by spin polarized hot electrons. Therefore, a legitimate and interesting question naturally arises: is it possible to demagnetize a ferromagnet using only unpolarized hot electrons and subsequently, how fast and efficient the effect can be? A first attempt to answer this question was reported by Eschenlohr *et al.* [27] where hot electrons were excited in a 30 nm thick Au layer deposited on top of a 15 nm Ni layer. It was concluded that hot electrons are as efficient as photons to demagnetize

Ni. However, the work has been questioned because of the strong overestimation of the light absorption in the Au layer [28], and in hindsight the structures were not optimal for demonstrating the effects looked for since a large photon absorption was present in the magnetic layer. More recently, a similar result was reported by Vodungbo *et al.* [29]. In that case, care was taken to avoid direct excitation of the ferromagnetic layer but all the conclusions are based on a single measurement. Furthermore, Eschenlohr's and Vodungbo's investigation were performed on magnetic samples much thicker than the hot electrons penetration depth while the experimental technique was sensitive to the whole thickness. Therefore, it is not obvious if the different features observed between laser and hot-electron induced dynamics are due to the process itself or to the inhomogeneous excitation.

In this paper, we aim at settling the controversy concerning the efficiency of ultrafast magnetization induced by hot electrons by presenting unambiguous experimental data of fast demagnetization induced by hot electrons only, proving a strong efficiency of the process and, demonstrating that it can be explained within a microscopic Three-temperatures model when adding hot electron ballistic transport.

The investigated samples consisting of Glass/Ta(3)/Pt(3)/$[Co(0.6)/Pt(1.1)]_2$/Co(0.6)/Cu(d)/Pt(3) (units in nm) were grown by dc magnetron sputtering, with d the Cu thickness varying from 0 to 300 nm. Cu was chosen since it has one of the highest hot electrons lifetime [17]. TR-MOKE experiments were realized using a pump-probe technique. The pump consists of 800 nm laser pulses, corresponding to a photon energy of 1.55 eV, with a 35 fs duration. The probe is frequency doubled to 400 nm. The laser repetition rate is 5 kHz. Both beams hit the sample at almost normal incidence and they are focused down to 300 μm and 60 μm for the pump and the probe respectively. Measurements were performed for both directions of the magnetization by pumping the sample from the top Pt/Cu face, while probing the Kerr effect through the glass substrate (Fig. 1a). In order to accurately determine the zero time delay, samples were patterned to have areas with and without the Cu layer.

In order to quantitatively separate direct photon excitation from the one due to hot electron transport, knowledge of the exact absorption profile in the multilayers is crucial. Therefore, the optical transmission in samples consisting of Glass/Ta(3)/Cu(d)/Pt(3) (units in nm) have been measured and compared to calculations based on absorption profiles [28]. Furthermore, absorption profiles were calculated for the full stack containing the [Co/Pt] multilayer. Because of the high absorption of Pt compared to Cu, the majority of the light is

absorbed in the top Pt layer (see supplementary information). Since its thickness is smaller than the hot electrons relaxation length at this energy, hot electrons generated in Pt can propagate to the Cu layer without experiencing significant scattering. As the Cu thickness increases, direct laser excitation of the Co/Pt multilayers is strongly reduced, since photon transmission decreases exponentially with the Cu thickness. The absorption in the Co/Pt multilayers is reduced by a factor of 100 for 60 nm of Cu, while hot electrons can pass such a layer almost without any loss.

In Fig. 1b, we show the measured ultrafast transient of the Kerr rotation for different thicknesses of the Cu layer. The laser fluence was adapted to obtain similar demagnetization (around 10 % except for 200 and 300 nm of Cu for which the maximum reachable demagnetization before damaging the sample was only 7.5 and 6% respectively) minimizing the effect of demagnetization amplitude on the dynamics [9]. It is therefore legitimate to normalize the data. First of all, a demagnetization is observed for Cu thicknesses as large as 300 nm. Since photons do not penetrate so far, an other source for demagnetization has to be considered. Then, a clear delay of the demagnetization onset is observed with increasing the Cu thickness. Finally, this delayed onset is accompanied by a more gradual, slower demagnetization, both to be quantified next.

From these measurements, we extracted the modification of the delay time in the onset of demagnetization ($\Delta t_0 = t_0(d) - t_0(d = 0)$) measured at 10% of the total signal, as close as possible of the start to minimize spurious effects, as well as the modification of the characteristic demagnetization time ($\Delta\tau(d) = \tau(d) - \tau(d = 0)$), both as function of the Cu thickness (Fig. 2). To do so, we fitted our M(t) magnetization traces using an empiric model given by $\frac{\Delta M}{M}(t) = \frac{A1-A2}{1+e^{\frac{t-(t_0+2.2\tau)}{\tau}}} + A2$ with A1 being the value at negative time delays and A2 corresponding to the value reached at maximum demagnetization, t_0 is the time corresponding to 10% of the maximum demagnetization and τ represents the characteristic demagnetization time. A linear evolution of the delay time with the Cu thickness is observed from 0 up to 300 nm of Cu. The slope corresponds to a constant velocity of $0.68 \times 10^6 \pm 0.05 \times 10^6$ m/s assuming ballistic trajectory perpendicular to the Cu layer. Such a linear variation and high velocity can only be attributed to hot electron transport. Indeed, the induced delay being within the hundreds of femtosecond range completely rules out the possibility of heat propagation by phonons since the sound velocity in Cu is about 4730 m/s which would result in a much longer delay. Moreover, the fact that the fit intercept

is close to 0 means that most of the hot electrons come from the top Cu/Pt interface, in agreement with the calculated large absorption and hot electrons generation in the top Pt layer (see supplementary information).

The linear variation of the time delay up to a 300 nm Cu thickness is characteristic of ballistic transport. Similar linear delay time in the transient reflectivity was observed in Au samples [30, 31]. Furthermore, the characteristic demagnetization time also shows a linear increase as a function of the Cu thickness, which could be understood as a broadening of the hot electron pulse (Fig. 2 (bottom)). This slower demagnetization will be at least partially caused by a spread in arrival times of the hot electrons, due to different (even almost random) propagation directions and different energies [32]. Note that the increase in demagnetization time is less pronounced than in the results reported by Vodungbo *et al.* [29]. There are two reasons for this difference. First, since the thin Pt layer is responsible for the generation of hot electrons, we expect a shorter hot electron pulse duration [33]. Second, the thickness of $[Co/Pt]_2$ in our samples is only 4 nm, compared to 18 nm in the case of Vodungbo *et al.* [29], meaning that we are far less sensitive to inhomogeneous excitations.

Insights in the hot electron transport properties and relaxation were obtained by measuring the evolution of the maximum demagnetization ($\Delta\theta_{max}$) as a function of the laser power for different Cu thicknesses (Fig. 3). The reference sample (i.e. d = 0 nm) for which there is a direct excitation of the Co/Pt shows first a linear increase at low power followed by a slowing down when approaching complete demagnetization, similarly to what has been reported before [35]. The same trend is observed for a Cu thickness of 20 nm. Above 40 nm, only the linear part can be observed since the maximum reachable demagnetization remains below 80%. The slope of maximum demagnetization as a function of the laser power decreases as the Cu thickness increases. Moreover, it should be pointed out that the demagnetization induced by hot electrons is very efficient and is as large as 40%, even for a Cu thickness of 120 nm. Therefore, similar behaviors are obtained for direct photon and hot electron excitations.

In order to study the decay of demagnetization efficiency more quantitatively, we plot the demagnetization at fixed laser power as a function of Cu spacer thickness, whereby clearly distinct regimes can be found. A separation between demagnetization due to direct photon excitation and hot electron excitation can be observed which occurs around 60 nm of Cu (areas I and II in Fig. 3b). For Cu layers thinner than 60 nm, the decay is rather fast cor-

responding to an exponential reduction of the contribution to the induced demagnetization of the direct photon absorption in the Cu layer. Above this thickness, we enter the second regime which could also be characterized by an exponential decay but with a much longer characteristic length of $150 - 200$ nm. In that case, demagnetization is only induced by hot electrons. Above 180 nm, a stronger attenuation seems to occur but the reason of this effect remains unclear and requires more experiments which is beyond the scope of this work. Moreover, the distinction between the direct photon and hot electron excitations vanishes when the pulse laser power increases. In that case, the direct absorption becomes sufficient to induce a large demagnetization even for a thicker Cu layer.

To investigate in more details the role of hot electrons in the ultrafast demagnetization process, we implemented hot electron ballistic transport within the Microscopic 3-temperatures model (M3TM) [9]. This model which was successfully used to reproduce laser induced magnetization dynamics for a wide range of ferromagnets, simulates transfer of angular momentum between the spin and the lattice based on Elliot-Yafet type of scattering with a probability a_{sf} that an electron flips its spin when absorbing or emitting a phonon (see reference [9] for more details). The time evolution of the electron and phonon temperature and of the magnetization is derived using a set of coupled differential equations given by:

$$\begin{aligned} C_e[T_e](z)\frac{dT_e(z)}{dt} &= g_{ep}(T_p(z) - T_e(z)) + P(z,t), \\ C_p\frac{dT_p(z)}{dt} &= g_{ep}(T_e(z) - T_p(z)), \\ \frac{dm(z)}{dt} &= Rm(z)\frac{T_p(z)}{T_C}\left(1 - m(z)\coth\left(\frac{m(z)T_C}{T_e(z)}\right)\right), \end{aligned} \tag{1}$$

where T_C is the Curie temperature of the ferromagnetic layer. T_e and T_p are the electron and phonon temperature respectively, and $P(z,t)$ represents the source of excitation as discussed below. Furthermore, $R = (8a_{\mathrm{sf}}g_{ep}T_C^2)/(k_B T_D^2 D_S)$ represents a prefactor controlling the demagnetization rate with k_B the Boltzmann constant, T_D the Debye temperature and D_S the atomic magnetic moment divided by μ_B. The coupling constant between the electron and phonon subsystem is given by $g_{ep} = (3\pi D_F^2 D_P k_B^2 T_D \lambda_{ep}^2)/(2\hbar)$ with D_F the density of states at the Fermi level, D_P the number of oscillators per atomic site and λ_{ep} the electron-phonon coupling constant. The parameters used for the simulations are adopted from ref. [37] (see supplementary information).

Transfer matrix calculations were performed to calculate the laser absorption profile for relevant Cu spacer thicknesses (see supplementary information). All absorption in the buried Co/Pt layer $A_{CoPt}(t)$ is treated as a direct heating source $P_{dir}(t)$ in Eq. (1), with a Gaussian temporal profile $exp(-(t/t_p)^2)$, where t_p is determined by the laser pulse length. All absorption in the top Pt and Cu, $A_{Pt}(t)$ and $A_{Cu}(t)$ resp., is assumed to be transferred to hot electrons. For sake of simplicity, and because of the short optical penetration depth, we approximate all hot electrons to be generated at t = 0 in a slab with infinitesimal thickness at the Pt/Cu interface. Hot electrons are assumed to travel ballistically at speed v_0, and with an isotropic orientational distribution, through the Cu of thickness d. Therefore, scattering that would widen the arrival time distribution is not taken into account. For this situation it is straightforward to derive the arrival time distribution at the Cu-Co/Pt interface. Furthermore we introduce a hot electron life time τ_{he} (related to the hot electron attenuation length via $\lambda_{he} = v_{he}/\tau_{he}$), and an efficiency F to transfer the energy flux at the interface into heating of the Co/Pt electronic system, which is treated as an indirect heating source term $P_{ind}(t)$ in Eq. (1). Thus we find:

$$P(t) = P_{dir}(t) + P_{ind}(t) \tag{2}$$

$$P_{ind}(t) = F(A_{Pt} + A_{Cu})(1 - \frac{d/v_{he}}{t^2})exp(-t/\tau_{he}), \text{if } t < d/v_{he}, \text{and 0 else} \tag{3}$$

Note that all $A_i(t)$ are a function of layer thickness, as explicitly calculated, but were approximated by simplifying analytical functions (suppl. information).

As observed in fig. 4a, the increase in the delay as a function of the Cu thickness is readily reproduced. In order to simulate the maximum demagnetization as a function of the power and the Cu thickness, we used a characteristic hot electron decay length of 200 nm, a hot electron velocity of 1×10^6 m/s and an efficiency parameter F of 0.8 (Fig. 4b). It should be noted that values between 0.7 and 1 gives qualitative agreement between experiments and simulations. The calculated evolution of the maximum demagnetization as a function of the Cu layer agrees very well with the experimental results. These results shed light on some of the questions that are still unanswered about ultrafast demagnetization. The first conclusion we can extract without ambiguity from this work is that the laser field is not a compulsory element to induce ultrafast demagnetization[2, 3, 36]. Moreover, in order to induce demagnetization of the buried Co/Pt multilayer, the hot electron energy has to be transferred into the

magnetic system. The generated unpolarized hot electrons in the Pt/Cu layers only act as a heating source, subsequently increasing the electron temperature by electron-electron interaction within the magnetic layer. We find very good agreement with experiments employing such a scenario and assuming a demagnetization process based on Elliot-Yafet mediated spin flip scattering, whereas in Eschenlohr's work [27], the hot electron induced demagnetization was claimed to be due to hot electron transport in the ferromagnetic layer itself. Despite the proven finite role of hot electron transport on ultrafast demagnetization, present consensus is that such demagnetization processes are predominantly driven by local dissipation of angular momentum [38]. Thereby we believe to have identified a more likely scenario for demagnetization induced by hot electron transport. In addition, the large value of the efficiency parameter F is quite surprising taking into account that hot electron propagation should be isotropic. This seems to show that this is not the case and that hot electrons preferentially propagate from the top excited surface to the bottom part of the sample. A similar conclusion was also reported by Turgut *et al.* [34].

Finally, further simulations based on heat diffusion due to electronic transport and removing the heat source due to hot electrons have been performed. In such a case, it is not possible to reproduce our results showing that heat diffusion, if present, would play only a minor role in the ultrafast demagnetization process (see supplementary information). Therefore, due to the large number of samples investigated in this work, we can assure that demagnetization is triggered by hot electrons.

Twenty years ago, a new field of research started when photons were shown to induce ultrafast demagnetization. In this paper we demonstrated unambiguously that not only photons but hot electrons can also induce ultrafast demagnetization in a very efficient way. The demagnetization dynamics is shown to be similar for hot electrons and direct laser excitations. Hot electrons could then be described as ballistic for Cu thickness up to 200 nm. This description added to a three Temperature Model can reproduce accurately our experimental results. Those findings open new perspectives. Indeed the role of hot electrons will have to be (re)considered carefully in the understanding of magnetization dynamic phenomena such as All Optical Magnetization Switching. Moreover engineered devices using hot electrons and spin filtering could be used to increase the efficiency of ultrafast spin transfer torque with the ultimate goal of controlling and manipulating the magnetization of a ferromagnetic thin layer.

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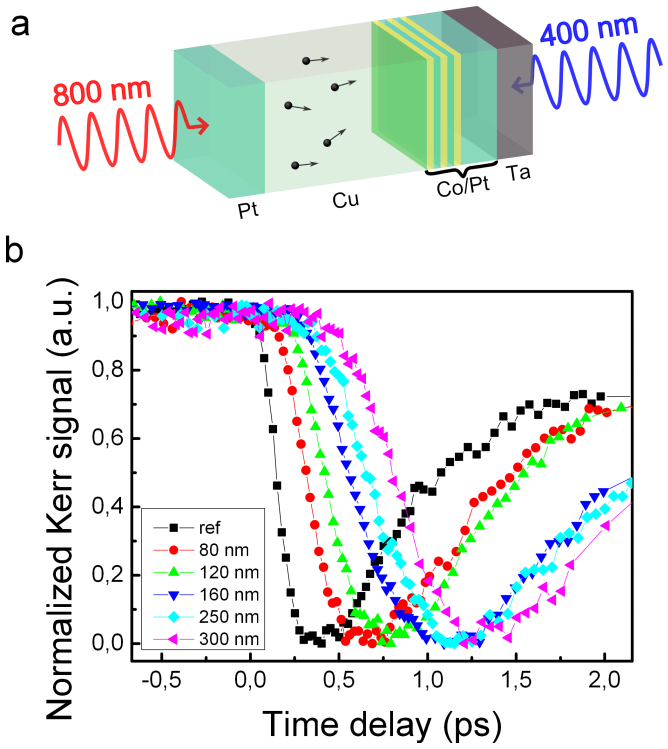

FIG. 1: (a) Schematic representation of the multilayer structure. (b) Time evolution of the magneto-optical signal measurements for different Cu thicknesses. The laser power was adjusted to keep the maximum demagnetization constant for all samples, allowing for a normalization of the signal.

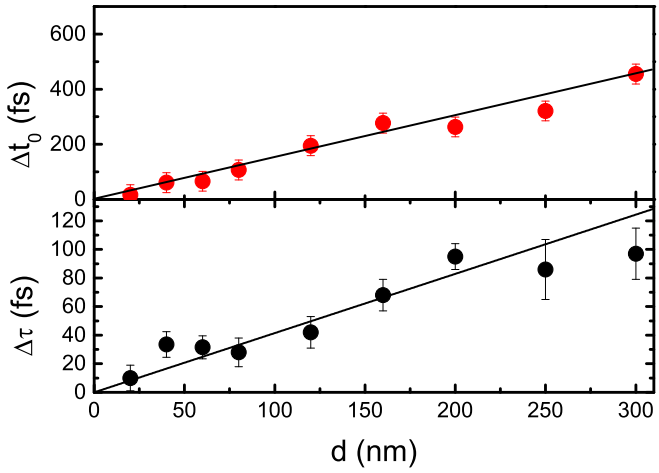

FIG. 2: (top) Cu thickness dependence of the induced time delay in the onset of demagnetization $\Delta t_0 = t_0(d) - t_0(d = 0)$. The full line is a linear fit to the data. (bottom) Modification of the characteristic demagnetization time $\Delta\tau(d) = \tau(d) - \tau(d = 0)$ as a function of the Cu thickness d. The full line is a linear fit to the data.

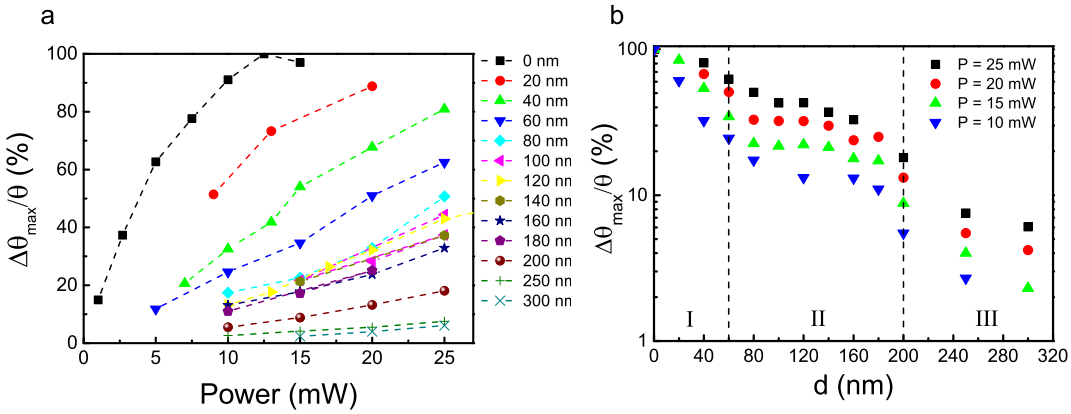

FIG. 3: (a) Power dependence of the induced Kerr rotation variation for different Cu Thicknesses *d*. (b) Thickness dependence of the induced Kerr rotation variation for different power. Zone I correspond to a combined direct photons and hot electrons excitations. In Zone II, only hot electrons are responsible for the demagnetization. Zone III corresponds to a faster attenuation of the demagnetization.

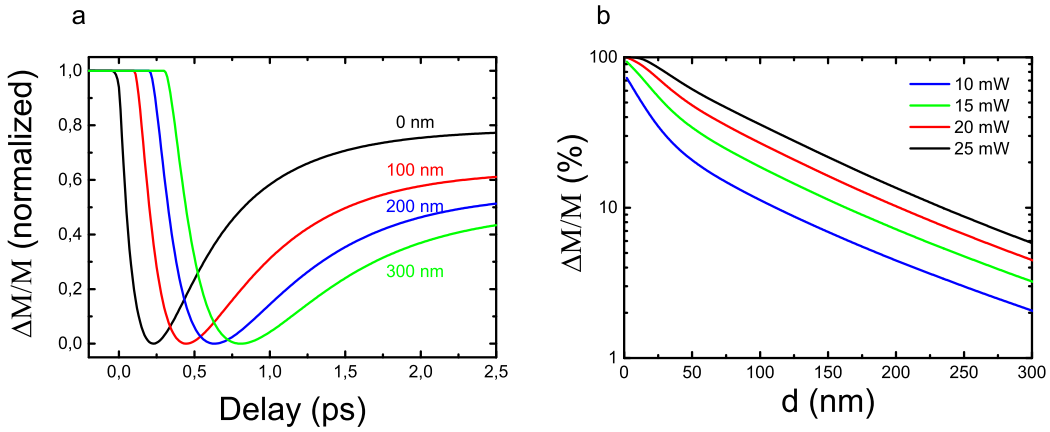

FIG. 4: (a) Simulation of ultrafast demagnetization for a laser power of 3.5 mW and different Cu thicknesses (b) Simulation of the maximum demagnetization as function of the Cu thickness for different laser power excitations. The hot electron attenuation length was set to $200nm$ (See supplementary information for details about the parameters used in the simulations).