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Published in:
Applied Physics Letters

DOI:
10.1063/1.4902069

Published: 17/11/2014

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Download date: 29. Dec. 2018
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Citation: Applied Physics Letters 105, 202402 (2014); doi: 10.1063/1.4902069
View online: http://dx.doi.org/10.1063/1.4902069
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Ultrafast magnetization dynamics in high perpendicular anisotropy [Co/Pt]n multilayers
Spin-orbit enhanced demagnetization rate in Co/Pt-multilayers

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(Received 2 October 2014; accepted 6 November 2014; published online 18 November 2014)

In order to explore the role of enhanced spin-orbit interactions on the laser-induced ultrafast magnetization dynamics, we performed a comparative study on cobalt thin films and Co/Pt multilayers. We show that the presence of the Co/Pt interfaces gives rise to a three-fold faster demagnetization upon femtosecond laser heating. Experimental data for a wide range of laser fluences are analyzed using the Microscopic 3-Temperature Model. We find that the Elliott-Yafet spin-flip scattering in the multilayer structure is increased by at least a factor of four with respect to the elementary Co film. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4902069]

Beaurepaire 1 showed in 1996 for the first time that the magnetization of a ferromagnetic transition metal can be quenched within a few hundred femtoseconds (fs) by an intense fs-laser pulse. Since these measurements, ultrafast laser induced magnetization dynamics has become a hot topic. The interest was not only caused by the new and exciting physics involving the demagnetization process, but also the appealing possibility of decreasing the magnetization switching time from the sub-nano- to the femtosecond time domain, 2 where recently exciting new fs laser-induced switching scenarios have been discovered using linearly 3- and circularly-polarized laser light. 4,5 So far, several models trying to unravel the processes were introduced; yet, the role of the individual subsystems (photons, electrons, phonons, and spins) in the ultrafast demagnetization process is heavily debated on. A central question is to understand how the angular momentum of the spin system is being transferred at a ~100 fs timescale during the demagnetization process. It is now widely believed that spin-orbit interactions play an important role as confirmed, e.g., by recent X-ray Magnetic Circular Dichroism (XMCD). 6,7 Therefore, tuning these spin-orbit interactions could provide an interesting route to tailor the magnetization dynamics to our own demands.

The energy flows among the electron, phonon, and spin subsystem induced by the intense laser pulse are qualitatively described by the phenomenological 3-Temperature Model (3TM). 1 The demagnetization traces as experimentally obtained are well reproduced by several recently proposed models addressing in more detail the underlying mechanism of the demagnetization process, such as the momentum-resolved Boltzmann scattering, 8-10 atomic Landau-Lifshitz-Gilbert (LLG), 11 Landau-Lifshitz-Bloch (LLB), 12 and the Microscopic 3-Temperature Model (M3TM). 13 The approach of the M3TM as compared to the atomistic LLG and LLB model is completely different. The latter two treat angular momentum transfer phenomenologically, using a damping parameter to account for the momentum transfer, whereas the M3TM is a microscopic model, treating individual scattering events quantum mechanically. It explicitly models transfer of angular momentum between the electron, lattice, and spin system by means of an Elliott-Yafet type of spin scattering characterized by the spin-flip probability $a_{sf}$. We are aware of alternative explanations for demagnetization via nonlocal mechanisms like the superdiffusive spin transport model as introduced by Battiato. 14 However, to explain our experimental results a local approach by the M3TM suffices.

So far, the M3TM has been successful at reproducing both the fast demagnetization traces of ferromagnetic transition metals such as Co and Ni 13,15 as well as Gd-materials exhibiting a relatively slow “two-step” demagnetization process. Moreover, Roth et al. 16 showed that Ni can exhibit a two-step demagnetization process by performing large laser fluence experiments at elevated environmental temperature as predicted by theory. 13

The minimum demagnetization time measured for all the elementary 3d ferromagnetic transition metals is typically 70–100 fs in the low fluence limit. In this letter, we examine the possibility to speed up this demagnetization process by manipulating the spin-orbit interactions. To achieve this, we make use of the strong spin-orbit coupling of Co at the interface of a Co/Pt-multilayer film. Due to the increased spin-orbit coupling we expect that Co/Pt has a decreased demagnetization time and increased $a_{sf}$ compared to ordinary Co. Using a TimeResolved Magneto Optical Kerr Effect (TRMOKE) setup, we performed laser-heating induced demagnetization measurements. Throughout the measurements, we varied the strength of the fs-laser pump pulse. These demagnetization traces are analyzed in the framework of the M3TM to obtain $a_{sf}$ of Co in this structure and the value is compared to its intrinsic bulk value.

In earlier work, 13 the M3TM was introduced and discussed in detail. Here we only recall its key features. The simplified model Hamiltonian of the M3TM describes spinless free electrons (e), phonons according to the Einstein or Debye model (p) and spin excitations obeying the mean-field Weiss model (s). For simplicity, the spin specific heat is set to zero and the electron gas is assumed to thermalize infinitely fast. For the electron specific heat $C_e$ we make the usual linear approximation of $C_e(T_e) = \gamma_e T_e$, with $\gamma_e$ a materials-dependent parameter and $T_e$ the electron temperature.
Furthermore, the phonon specific heat \( C_p \) is proposed to be independent of the phonon temperature \( T_p \). We also assume that the heat diffusion is dominated by the electrons and described by the heat conductivity \( \kappa \). Realizing that the investigated layers are much thinner than the probed region, it is justified to limit the model to one dimension only, i.e., the \( z \)-coordinate relative to the surface of the ferromagnet. As a result, we can derive a set of coupled differential equations for the electron and lattice temperature:

\[
C_e(T_e(z)) \frac{dT_e(z)}{dt} = \nabla_z (\kappa \nabla_z T_e(z)) + g_{ep}(T_p(z) - T_e(z)) - P_{pump}(z,t); \\
C_p \frac{dT_p(z)}{dt} = g_{ep}(T_e(z) - T_p(z)); \\
\frac{dm(z)}{dt} = Rm(z) \left( \frac{T_p(z)}{T_C} \left[ 1 - m(z) \coth \left( \frac{mT_C}{T_e(z)} \right) \right] \right),
\]

in which \( g_{ep} \) represents the coupling constant between the electron and phonon subsystem, \( m = \frac{m}{M_s} \), the magnetization relative to the saturation value at \( T = 0 \) (\( M_s \)) and \( T_C \) the Curie temperature of the ferromagnet. The prefactor \( R \) is a rate (dimension \( s^{-1} \)) and equals \( R = (8a_0^2 \gamma C_{ep})/(k_B T_D D_r) \), with \( T_D \) the Debye temperature and \( D_r \) the atomic magnetic moment divided by \( \mu_B \). Furthermore, we use the expression for \( g_{ep} = (3\pi D_r^2 \rho B_0 T_D^2 \gamma^2)/2h \), with \( D_r \) the density of states around the Fermi-level, \( \rho \) the amount of possible polarization states and \( \gamma \) the electron-phonon coupling constant. Finally, the fluence of the laser pump pulse is included in the model under the assumption that it is fully absorbed by the electron system. To satisfy the finite penetration depth and the Gaussian time-profile of the laser pulse as well, the source term \( P_{pump} \) in the expression for the electron temperature is written as \( P_{pump}(z,t) = \frac{P_0}{\sqrt{\pi} \sigma} \exp \left( -\frac{z^2}{\sigma^2} \right) \), in which \( P_0 \) is the total fluence of the laser pulse, \( \sigma \) is the penetration depth, and \( \lambda \) describes the laser pulse length.

The Co/Pt-multilayer was fabricated by DC magnetron sputtering on a boron doped Si substrate with native oxide on top. Its exact layout is Pt5/(Co0.5Pt0.6)11/Pt1.4 (units in nm). We emphasize that under our deposition conditions non-negligible inter-mixing takes place at the interfaces, and that the samples have out-of-plane anisotropy. Two series of TRMOKE measurements were carried out on this sample. The first series is optimized for high fluences of the laser pump pulse and thereby large quenching of the magnetization. The pump pulses are linearly polarized and their wavelength is centered around 800 nm, whereas the probe pulse has a central wavelength of 400 nm. Furthermore, the repetition rate of the laser pulses is 1 kHz, the pulse length is characterized by \( \sigma = 35 \) fs, and the spot size of laser pulse on the sample is typically 3 mm for the pump and 0.2 mm for the probe pulse. In the second series, the fluence of the laser pump pulse is considerably lower than the first series. The wavelength of both the pump as well as the probe pulse is centered around 790 nm with a pulse width characterized by \( \sigma = 45 \) fs and a repetition rate of 80 MHz. In the high fluence series, the laser fluence was varied by changing the power of the pump pulse itself. In order to change the fluence in the low fluence series, however, the power of the pump was kept constant, but the spot size was decreased from 8 to 4 \( \mu \)m by substituting the laser objective. TRMOKE data of an ordinary 15 nm Co film dc-sputtered on a MgO-substrate is used as reference.

Figures 1(a) and 1(b) show the results of the high and low fluence TRMOKE measurements carried out on the Co/Pt-multilayer, displaying the magnetization \( M(t) \) normalized to the magnetization at the temperature just before the pump pulse excitation \( M_0 \). In Figure 1(c) the TRMOKE data from the Co film is presented for comparison. The shape of all the TRMOKE curves in the figure is characteristic for ferromagnetic transition metals: first an ultrafast (fs) quenching of the magnetization, or demagnetization, appears directly from the moment the pump pulse arrives, followed by a slower (ps) remagnetization process. Furthermore, the maximum demagnetization \( (\Delta M_{max} M_0) \) increases for larger fluences, and its position shifts to larger delay times. However, we can also directly observe a clear difference between the data from the Co/Pt-multilayer and Co sample: the time needed to reach the maximum demagnetization for the Co/Pt multilayer is significantly shorter than for the Co sample with the same maximum quenching.

To quantify this observation, we extract a value of the demagnetization time for each magnetization trace in Figure 1, using the approach of Dalla Longa to parameterize the traces. Our procedure yields the experimental decay time \( \tau_{fr} \)
defined as the time needed for the magnetization to reach a level of \((1 - e^{-1})\) of its maximum demagnetization corrected for the finite pulse length of the laser. The individual data points in Figure 2 represent the experimental values of \(\tau_M^*\). From the figure, it becomes clear that \(\tau_M^*\) for Co/Pt is generally about 2–3 times smaller, e.g., for \(\Delta M_{\text{max}}/M_0 = 0.5\) we determined \(\tau_M^*\) to be about 240 fs for Co and only 90 fs for Co/Pt. Furthermore \(\tau_M^*\) generally increases for increasing fluence for both samples.

As we want to extract microscopical information from the data in Figure 1, we set up a simulation based on the M3TM characterized by Eq. (1) for both the Co/Pt as well as the ordinary Co film. The key materials in these simulations are the pseudo-material “Co/Pt” for the Co/Pt-multilayer and Co for the reference film. Co and Pt are thus put together and are the pseudo-material “Co/Pt” for the Co/Pt-multilayer and the ordinary Co film. The key materials in these simulations are the pseudo-material “Co/Pt” as in Ref. 16 it is stated that an overestimation leads to an increased value for \(\alpha_{sf}\). To verify this, we evaluate two scenarios for this parameter. In the first scenario, all five parameters including \(C_i/C_p\) are unconstrained to reproduce the data, whereas in the second scenario, \(C_i/C_p\) is fixed according to the weighted average based on the literature values. 18 For the data of the Co reference sample, we only performed the analysis according to the second scenario.

The solid lines in Figure 1(a) represent the results of the fit procedure according to the first scenario for the Co/Pt-multilayer. Analogously the lines in Figure 1(c) result from the fit procedure for the Co reference film. The values of the fit parameters are listed in the second and fourth column of Table I. Figures 1(a) and 1(c) show that there is a good agreement between the M3TM simulation and the demagnetization traces from the TRMOKE measurements. From the fit values, we extracted the average spin-flip probability \(a_{sf} = (10.5 \pm 0.5) \times 10^{-2}\) for “Co/Pt” and \(a_{sf} = (2.5 \pm 0.5) \times 10^{-2}\) for Co in the reference sample. So, the average spin-flip probability of “Co/Pt” has increased by a factor of four as compared to normal Co. As the main difference between the multilayer and reference sample relies on adding Pt-layers, which increases the spin-orbit coupling, we conjecture that the increased value of \(a_{sf}\) is a result of the increased spin-orbit coupling. The values of \(\Delta \varphi_p\), \(C_i/C_p\) and \(T_C\) for “Co/Pt” and Co are in line with the numbers as reported by Lin et al.,19 Roth et al.,16 and van Kesteren and Zeper.20 However, \(\kappa\) for “Co/Pt” and Co are only a fraction of what we would expect from their individual values, being 100 and 71.6 J s\(^{-1}\) m\(^{-3}\) K\(^{-1}\). One may speculate that the reduced heat conductivity is due to many Co-Pt interfaces in the multilayer structure, finite size effects and short timescales and/or the highly non-uniform heating of the sample. A more detailed analysis of \(\kappa\) is certainly of interest but beyond the scope of the present work, as it is not of particular importance for the here presented results.

In the second scenario, we fix \(C_i/C_p\) to what is expected from the literature values of \(\gamma_{Co} = 6.65 \times 10^2\) and \(\gamma_{Pt} = 7.19 \times 10^2\) J m\(^{-3}\) K\(^{-1}\). The resulting demagnetization traces are not shown in Figure 1(a) as they are hardly resolvable from the traces of the first scenario. The data in column 3 of Table I show that \(\Delta \varphi_p\) is comparable to the value for Ni as determined by Roth16 and almost equal to the value of Co in the reference sample. The value of \(T_C\) becomes smaller, but still is reasonable and the number for \(\kappa\) increases considerably; yet, it remains significantly smaller than its weighted average value as expected. More importantly, \(a_{sf}\) increases to \((13.5 \pm 0.5) \times 10^{-2}\) being five times our value of Co. Finally, we note that the value of \(a_{sf}\) for the Co reference sample is much smaller than reported in Ref. 13. This observation is in line with the findings of Roth for Ni. They corrected the earlier reported \(a_{sf}\) for Ni from \(18.5 \times 10^{-2}\) (Ref. 13) to \(8 \times 10^{-2}\) and attributed this to an overestimation of \(C_i\), relatively to the total heat capacity of Ni in Ref. 13 leading to a larger spin-flip probability. We see that an increased \(C_i/C_p\) in scenario 2 indeed leads to an increased value of \(a_{sf}\) relative to scenario 1. We conclude that the exact value of \(a_{sf}\) is dependent on the choices made for the other material parameters, but also that, independent of these choices, \(a_{sf}\) is found to be significantly larger for the Co/Pt structure as compared to the Co reference film.

As the demagnetization process develops faster at low laser fluences, we performed low fluence TRMOKE measurements on a different setup. The values of \(\tau_M^*\) extracted from the two curves in Figure 1(b) are plotted in Figure 2.

![FIG. 2. The demagnetization times determined from the individual traces of Figure 1 using the approach of Ref. 17. The dashed lines represent the results from simulations based on the M3TM.](image-url)
and are both smaller than the smallest \( \tau_M \) of the high fluence series of measurements. More precisely, the smallest \( \tau_M \) is only 40 fs. To check the consistency of the fit parameter values as determined from the high fluence data according to scenario 1, we perform a fit routine on these low fluence data. As the range of maximum quenching of the magnetization is limited, we have to add constraints in order not to over-parameterize the problem. Therefore, the parameters \( T_C \) and \( \kappa \) are fixed to the values as determined in scenario 1. The solid lines in Figure 1(b) represent the results of the global fit routine carried out on the two demagnetization traces. The simulation reproduces the experimental data well. The magnitudes of the “Co/Pt” parameters of interest equal \( a_{sf} = (10.2 \pm 0.5) \cdot 10^{-2} \), \( C_d/C_p = (2.5 \pm 0.3) \cdot 10^{-2} \) and \( \lambda_{ep} = (55 \pm 5) \) meV, and thus all of them are in agreement with the values of scenario 1. We therefore have shown that \( a_{sf}, C_d/C_p \) and \( \lambda_{ep} \) display no pronounced dependency on the laser fluence and that the values extracted are consistent among the two different TRMOKE setups used in the experiments. Because all the properties of both samples are well determined by means of our M3TM simulations, we can make a comparison between \( \tau_M \) as determined experimentally and by the M3TM. The values for \( \tau_M \) originating from M3TM traces are represented by the dashed lines in Figure 2. The M3TM reproduces the trend of the demagnetization as a function of the magnetization quenching well for both samples, providing further confidence in the applicability of the M3TM for analyzing laser-induced demagnetization dynamics.

Finally, we try putting the (at least) four-fold increase in \( a_{sf} \) for the Co/Pt-multilayer in a more physical perspective. We consider this increase quite modest and realistic. Elliott-Yafet spin-flip scattering generally scales as \( Z^2 \), with \( Z \) the atomic number. As the atomic number of Pt is about three times as large as Co, this would lead to an enhancement of almost two orders of magnitudes. Even taking an atomic average throughout the Co/Pt-multilayer, and accounting for the fact that for an ideal interface the average Co atom only sees a few Pt neighbors, a significant enhancement for Co at a Co/Pt interface with respect to pristine Co could be easily achieved. Nevertheless, it should be emphasized that a more quantitative estimate is very challenging. A more serious calculation of the spin-flip probability for the Co/Pt structure would be highly interesting, but such an analysis goes well beyond the scope of our present letter.

In conclusion, we found that the M3TM can reproduce the resulting TRMOKE demagnetization traces of a Co/Pt-multilayer and the Co reference sample to great detail. We were able to describe all data on each sample by a single value of \( a_{sf} \) independent of the laser fluence. From this analysis, we conclude that the spin-flip scattering of Co in our multilayered structure is at least four times as large as that of ordinary Co, causing to decrease the demagnetization time considerably. These findings are in line with our conjecture that the enhanced spin-orbit coupling leads to a faster demagnetization process.

This work is part of the research programme of the Foundation for Fundamental Research on Matter (FOM), which is part of the Netherlands Organisation for Scientific Research (NWO).