

Laser induced ultrafast 3d and 4f spin dynamics in CoDy ferrimagnetic alloys as a function of temperature

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1	Title: Laser induced ultrafast 3d and 4f spin dynamics in CoDy ferrimagnetic alloys as a
2	function of temperature.
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20	
21	0. Abstract
22	
23	We report an element- and time-resolved investigation of femtosecond laser induced
24	ultrafast dynamics of the Co 3d and Dy 4f spins in a ferrimagnetic Co ₈₀ Dy ₂₀ alloy as a function
25	of the temperature. We observe that the Co characteristic demagnetization time (τ_{Co}) remains
26	nearly constant (~0.2 ps) on increasing the temperature. Conversely, the Dy characteristic
27	demagnetization time (τ_{Dy}) decreases from ~1 ps to ~0.4 ps with the rise of temperature.
28	Comparing our experimental data with literature shows that τ_{Co} and τ_{Dy} are independent of the
29	alloy composition or the demagnetization amplitude and that τ_{Dy} scales with the relative
30	temperature $T^* = T_{Curie} - T$.
31	
32	<u>Key words</u> :

Ferrimagnetic alloys, ultrafast laser induced demagnetization, femtosecond laser, time resolved X-ray Magnetic Circular Dichroism.

- 35 <u>Highlights</u>:
- Element- and time-resolved investigation of laser induced ultrafast Co3d and Dy4f spin
 dynamics in a ferrimagnetic CoDy amorphous alloy as a function of temperature.
- 38

The Co characteristic demagnetization time remains nearly constant (~0.2 ps) on
 increasing the temperature while the Dy characteristic demagnetization time decreases
 from ~1 ps to ~0.4 ps with the rise of temperature.

- 42
- 43 **1.** Introduction
- 44

45 Excitation of ferromagnetic layers with infra-red (IR) femtosecond (fs) laser pulses leads to a quenching of the magnetic order on a sub-picosecond time scale [1]. The microscopic 46 47 mechanisms governing this ultrafast demagnetization are still subject to controversy in spite of 48 intensive experimental and theoretical studies [2 - 8]. Although these investigations have 49 revealed fundamental dissimilarities in the laser induced ultrafast magnetization dynamics in 50 transition metals (TM) [1, 9 - 11] and in rare-earth (RE) films [12 - 16], a few general features 51 were established. For instance, a rise in temperature results in an increase of the characteristic 52 quenching times (τ) of the 3d magnetic order in pure transition metals (TM) and 5d magnetic 53 order in pure rare-earth (RE) films [11, 13, 17]. In the specific case of pure RE layers, such 54 behaviour is also expected for the localized 4f spins as theoretically predicted [13] and sustained 55 by experiments that have evidenced concomitant laser induced dynamics of the 5d and 4f spins 56 [12 - 14, 16, 18]. In these experiments, the IR fs laser pulses excite the RE 5d spins while the 57 RE 4f spin dynamics is indirectly triggered via the 5d-4f intra-atomic exchange coupling [12, 58 14].

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60 In RE-TM ferrimagnetic alloys, the RE 4f spin order originates from the Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange coupling [19]. The RE 4f – RE 4f indirect exchange coupling 61 62 is mediated by the RE 5d as well as the TM 3d electrons in the conduction band [20]. 63 Interestingly, the published element- and time-resolved experiments have reported distinct laser 64 induced ultrafast dynamics for the TM 3d and RE 4f spins in spite of this RKKY exchange coupling [21 – 30]. Recently, the Landau-Lifshitz-Bloch (LLB) model [13, 17] was extended 65 66 to treat the laser induced ultrafast dynamics in multi-sublattices ferrimagnetic alloys [31] such as FeCoGd [32], CoTb [33] and FeTb [34] alloys. The calculations based on this modified LLB 67

68 model have shown that the characteristic demagnetization times of both the FeCo (τ_{FeCo}) and 69 the Gd (τ_{Gd}) sublattices in FeCoGd alloys strongly depend on the difference between the initial temperature (T) and the Curie temperature (T_{Curie}) of the alloy [32]. This theoretical work 70 71 highlights explicitly the differences between the ultrafast laser induced TM 3d and the RE 4f 72 spin dynamics in these alloys with the variation of the temperature [32]. Hennecke et al. have 73 invoked the effect of temperature to explain the short Gd demagnetization time they have 74 evidenced in a FeCoGd alloy [30]. Ferté et al. have earlier investigated the laser induced demagnetization in Co₈₀Dy₂₀ and Co₇₈Dy₂₂ alloys at different initial temperatures but ensuring 75 76 a constant relative temperature T_{Curie} - T [27]. They have shown that the response of the Dy 77 (resp. Co) magnetization to laser excitation was the same for both alloys in line with the LLB 78 calculations. The distinct dynamics of 3d and 4f spins is believed to be the key ingredient for 79 ultrafast all optical switching [21, 35] as well as ultrafast spin-transfer torque assisted switching 80 [36] in RE-TM alloys. Thus, it is of paramount importance to determine the correlation between 81 the characteristic demagnetization times and physical parameters such as the temperature. 82 However, systematic element- and time-resolved investigations of laser induced spin dynamics 83 in a single RE-TM alloy as a function of temperature are still lacking.

84

85 In this work, we have studied the laser induced ultrafast dynamics of Co3d and Dy4f spins 86 in a Co₈₀Dy₂₀ ferrimagnetic alloy as a function of temperature by mean of time-resolved X-Ray 87 Magnetic Circular Dichroism (tr-XMCD) [37]. Interestingly, we report experimental evidences 88 that the dependence of the laser induced dynamics of Co 3d and Dy 4f spins on temperature are 89 clearly different. We observe that τ_{Co} remains nearly constant (~0.2 ps) while τ_{Dv} decreases from 90 ~1 ps to ~0.4 ps when the temperature rises from 160K to 350K. Furthermore, a comparison of 91 our experimental data with existing data from literature on laser induced ultrafast dynamics of 92 Dy 4f spins in CoDy alloys [25, 27] suggests that τ_{Dy} is determined by T* = T_{Curie} – T.

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2. Material and methods

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The 18 nm thick $Co_{80}Dy_{20}$ alloy layer was deposited by DC magnetron sputtering on a "heat sink" Ta(3)/Cu(20)/Ta(3) multilayer sustained by a Si₃N₄ membrane. The alloy was capped with a Al(3)/Ta(3) bi-layer to prevent oxidation. The $Co_{80}Dy_{20}$ alloy displays an out-of-plane magnetic anisotropy. We have recorded hysteresis loops at various temperatures using SQUID magnetometry to extract the dependence of the coercive field (H_C) on temperature. For these 101 measurements, we have used a test $Co_{80}Dy_{20}$ alloy layer deposited simultaneously with the one 102 used for the time-resolved experiments but on a Si substrate (figure 1). This figure shows a 103 divergence of H_C in the vicinity of T ~ 250K indicating the temperature of magnetic 104 compensation [38, 39].

105

106 The tr-XMCD experiments were carried out at the femtoslicing beam line of the BESSY II 107 synchrotron radiation source at the Helmholtz-Zentrum Berlin [37]. The magnetization 108 dynamics have been measured by monitoring the transmission of circularly polarized X-ray 109 pulses tuned to specific core level absorption edges as a function of a pump-probe delay for two 110 opposite directions of the magnetic field. The photon energy was set to the CoL₃ and the DyM₅ 111 edges using a reflection zone plate monochromator on UE56/1-ZPM. The full width at half 112 maximum (FWHM) of the 800nm pump laser was set to 500 µm to ensure homogeneous 113 pumping over the probed area of the sample (FWHM ~ $200 \mu m$). A magnetic field of $\pm 0.55 T$ 114 was applied along the propagation axis of both the IR laser and the X-ray beam during the experiment. The measurements were carried out at $T^* = 350$ K (configuration 1), 400 K 115 116 (configuration 2) and 540 K (configuration 3) with $T^* = T_{Curie} - (T_{cryo} + \Delta T)$ as illustrated in 117 figure 2. Here, T_{crvo} is the temperature of the cryostat and ΔT is the temperature elevation due 118 to the continuous laser heating (table 1). The Curie temperature of the $Co_{80}Dy_{20}$ alloy (T_{Curie} = 119 700 K) is extrapolated from literature [40] and from mean field calculations [41, 42]. As only 120 one single sample was used in this experiment, any small error on the estimated value of T_{Curie} 121 would shift T* without affecting our conclusions. We have determined that the coercive field 122 of the Co₈₀Dy₂₀ alloy was below 0.55 T either below 160 K or above 300 K by monitoring the 123 XMCD amplitude as a function of temperature. The divergence of H_C between 160 K and 300 124 K is related to $T_{comp} \sim 250$ K as illustrated in figure 1. As a consequence, the experimental 125 parameters, such as the pump laser powers (P) and T_{cryo} , have been chosen so that $T_{cryo} + \Delta T$ stay in the temperature ranges that allow for magnetic saturation of our alloy. We have relied 126 127 on the thermal variation of the coercive field to determine ΔT for the two different pump laser 128 powers (P = 17 and 50 mW) that we have used during the experiment. To do so, we initially set 129 $T_{cryo} = 80$ K and turned on the laser. P = 17 mW was the largest laser power for which T_{cryo} + ΔT stays below T = 160K. Above this temperature, the CoDy alloy could not be saturated. As a 130 131 consequence, we estimated that $\Delta T \sim 80$ K for P = 17mW. In order to estimate ΔT with P = 50 132 mW, we compared the hysteresis loops at $T_{cryo} = 300$ and 320 K with P = 0 mW to the hysteresis 133 loops recorded at negative delay for $T_{crvo} = 80K$ with P = 50 mW (figure 3). The same signs of 134 the hysteresis loops indicate that these measurements were performed above T_{comp}. Moreover,

we notice that the coercive field is smaller for P = 50mW and $T_{cryo} = 80$ K compared to P = 0135 136 mW and T_{cryo} = 300 and 320 K. According to the thermal variation of the saturation field above 137 T_{comp} (figure 1), we estimated that $T_{cryo} + \Delta T$ is above 320K for P = 50mW and $T_{cryo} = 80K$. 138 The shape of the hysteresis loop at P = 50 mW and $T_{cryo} = 80$ K also indicates that we are close 139 to the temperature of spin reorientation transition [43]. As a consequence, we estimated $\Delta T >$ 140 250 K for P = 50 mW. Our procedure to estimate ΔT results in significant error bars on T* 141 (figure 5). Nevertheless, we estimated that we performed the time-resolved experiments at T* 142 = 350 K, 400 K and 540 K (figure 2, table 1). The measurements were carried out above T_{comp} 143 for the configurations 1 and 2 (figure 2a and b) and below T_{comp} for the configurations 3 (figure 144 2c) [27].

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3. Experimental results and discussion

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148 The normalized transient XMCD signals recorded at the Co L_3 and Dy M_5 edges for $T^* =$ 149 350, 400 and 540 K are displayed in figure 4a, b and c respectively. At T*=540K (figure 4c), 150 the maximum demagnetization of the Co sublattice is reached while the demagnetization of the 151 Dy sublattice has barely started as reported in a large number of element- and time-resolved experiments for different RE-TM alloys [21 - 27]. In contrast, at T^{*} = 350K (figure 4a), the 152 153 magnetization of the Dy sublattice is close to its minimum value while the minimum 154 magnetization of the Co sublattice is reached. The tr-XMCD curves at the Co L₃ edges were 155 fitted with two exponential functions (respectively the demagnetization and the magnetization 156 recovery) convoluted with a Gaussian function which account for the experimental time 157 resolution (130 fs) [24, 44, 45]. It is worth noticing that we have imposed a lower limit at 130 158 fs for the characteristic demagnetization times during the fitting procedure. It means that the 159 actual τ_{Co} is possibly below the experimental time resolution for T* = 400 K and 540K. The tr-160 XMCD curves at the Dy M₅ edge were adjusted by a single exponential decay convoluted by a 161 Gaussian function since we did not observe any recovery on the recorded time range. We have 162 extracted the characteristic demagnetization times (τ) from these fits as well as their error bars, 163 which correspond to one standard deviation (table 1). The dependence of the characteristic 164 demagnetization times on temperature for both sublattices are displayed in figure 5. We also 165 report the characteristic demagnetization times for the Co and Dy sublattices in various CoDy 166 alloys measured by Ferté et al (in Co₈₀Dy₂₀ and Co₇₈Dy₂₂) [27] and Radu et al (in Co₈₃Dy₁₇) 167 [25] in figure 6. Ferté et al have explicitly given the numerical values for T_{cryo} , T_{Curie} and ΔT

168 [27] and therefore we have derived $T^* = 430$ and 450K for the Co₈₀Dy₂₀ and Co₇₈Dy₂₂ alloys 169 respectively. Radu et al have performed their measurements at T = 100K without considering 170 any DC heating. Therefore, we assume $\Delta T = 0$ K and we include an extended error bar for this 171 data. It is worth noticing that including a temperature elevation of $\Delta T \sim 100$ -200 K (typical 172 values estimated for the pump-probe experiments on thin films) will not change our main 173 message. Thus, we derived T*=930K since T_{Curie} = 1030K for their Co₈₃Dy₁₇ alloys [43].

- 174
- 175 *Table 1:* Characteristic demagnetization times extracted from the fit functions as a function of
- 176 temperature T*. The cryostat temperature (T_{cryo}) , the laser continuous heating (ΔT) , the laser

$T^{*}(K)$	T _{Cryo}	$\Delta T(K)$	Laser	Edge	Demagnetization
	(K)		power		time τ (fs)
			(mW)		
540	80	80	17	Co L ₃	130 ± 100
540	80	80	17	Dy M ₅	980 ± 200
400	220	80	17	Co L ₃	130 ± 60
400	220	80	17	Dy M ₅	570 ± 90
350	80	> 250	50	Co L ₃	212 ± 25
350	80	> 250	50	Dy M ₅	400 ± 100

177 *power and the X-ray absorption edges are also recalled.*

178

179 In figure 5, we observe that τ_{C_0} is almost constant within the error bars between T^{*} = 180 350 K and $T^* = 540$ K in spite of the various laser powers used, and thus the different 181 demagnetization amplitudes, in line with previous work by Jal et al [46]. We also observe a 182 clear decrease of τ_{Dv} when T* decreases from 540K to 350K (figure 5). In pure Gd layers, the 183 characteristic demagnetization time τ_{Gd} related to the (5d, 6s) magnetic order increases when 184 the laser power is increased [13, 16]. The concomitant quenching of itinerant (5d, 6s) and 185 localized 4f magnetic order in pure RE layers [12 - 16, 18, 47] suggests that such an increase is 186 also expected for the 4f spins. However, such a behavior is not observed in our CoDy alloy 187 since the shorter demagnetization time (τ_{Dv}) is obtained for the larger laser power (P = 50mW) 188 and thus also for the larger demagnetization amplitude. Therefore, we can rule out the distinct 189 laser power as the origin of the measured variation of τ_{Dy} with temperature. Gang et al. have 190 reported a decrease of the Ni 3d characteristic demagnetization time in NiPd ferromagnetic

191 alloys when the Curie temperature is reduced (and thus T*) by increasing the Pd concentration 192 [48]. They have attributed such feature to an increase of the spin-flip scattering probability [2] 193 with Pd content, caused by its larger spin-orbit coupling compared to pure Ni. In our case, this 194 explanation does not hold since we have studied a single alloy composition. According to the 195 LLB calculations in ferrimagnets [32], by increasing the temperature, we would expect to go 196 from a situation in which $\tau_{TM} < \tau_{RE}$ at low temperatures (T << T_{Curie}) to a situation in which τ_{TM} 197 ~ τ_{RE} at higher temperatures (T > 0.8 T_{Curie}). The transition between these dynamical regimes 198 requires that τ_{Dy} decreases and/or τ_{Co} increases in the intermediate temperature range. Our 199 experimental findings are thus consistent with this LLB prediction but call for further 200 experiments at even higher temperatures, especially in the vicinity of T_{Curie} to challenge further 201 their predictions.

202

203 Two qualitative descriptions can be proposed to explain the different temperature 204 dependent evolution of τ_{Co} and τ_{Dv} . The first one considers that different microscopic 205 mechanisms are supposed to be responsible for the laser induced ultrafast quenching of Co 3d 206 and Dy 4f magnetic orders. Indeed, the dynamics of TM 3d spins is presumably caused by spin-207 flip scattering [49 - 51] and superdiffusive spin transport [3, 52] while the dynamics of the RE 208 4f spins is claimed to be related to spin-waves [53 - 55]. The second one considers that the Co 209 3d spin dynamics is mainly governed by the direct ferromagnetic Co – Co exchange coupling 210 while the Dy 4f spin dynamics is mainly governed by the indirect antiferromagnetic Co – Dy 211 exchange coupling. Recent measurements have shown that antiferromagnetic and 212 ferromagnetic materials exhibit distinct laser induced ultrafast dynamics [56].

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214 Finally, in figure 6, we compare our experimental results with existing data from the 215 literature [31, 33] by plotting τ_{Co} and τ_{Dy} as a function of T* for different alloy compositions. 216 We observe that τ_{C_0} and τ_{D_V} are both constant (within error bars) from T* = 930 K to T* = 540 217 K. From T^{*} = 540 K to T^{*} = 350 K, we observe that τ_{Co} is constant while τ_{Dv} decreases, 218 confirming our experimental results from figure 5. It is also very interesting to note that the τ_{Dy} 219 extracted from the work of Ferté et al. are consistent with our present results for similar T* 220 although in their case the demagnetization amplitudes were larger than 80%. It suggests that τ_{Dy} does not dependent on the demagnetization amplitude but mainly on T* = T_{Curie} – T. 221

222

223 4. Conclusions

225 We have investigated the laser induced ultrafast dynamics of Co 3d and Dy 4f spins in a 226 ferrimagnetic CoDy alloy as a function of temperature by element- and time-resolved XMCD. 227 We have revealed striking differences between the Co 3d and Dy 4f spin dynamics when the 228 temperature is varying from 160 K to 350K. The characteristic demagnetization time of the 229 Dy4f sublattice decreases while it is almost constant for the Co3d sublattice. This experimental 230 findings sustain some of the predictions of the LLB model, namely that the characteristic 231 demagnetization time of the RE sublattice should be smaller than that of the TM sublattice at 232 high temperature. Our experimental results also confirm that the characteristic demagnetization 233 time of the Co sublattice does not depend on the composition, on the demagnetization amplitude 234 nor on the temperature in CoDy alloys as reported by Jal et al [46]. Finally, our data set, 235 amended with data extracted from literature, suggest that the characteristic demagnetization 236 time of the Dy sublattice is determined by $T^* = T_{Curie} - T$ and does not depend on the 237 demagnetization amplitude or alloy composition. Our work calls for further experimental 238 investigations at elevated temperatures to challenge the predictions of the LLB model for spin 239 dynamics in RE-TM alloys, especially in the vicinity of T_{Curie} . Such experimental confirmation 240 would link the characteristics of laser induced ultrafast dynamics and the static magnetic 241 properties of ferrimagnetic alloys [32]. We hope this work will motivate further experimental 242 investigation at elevated temperatures as well as development of the LLB model to 243 ferrimagnetic alloys [57].

244 245 Figures:





248 Figure 1: Dependence of the coercive field (H_c) on the temperature for a $Co_{80}Dy_{20}$ alloy measured by means of 249 SQUID magnetometry (red empty circles). H_C diverges in the vicinity of $T \sim 250K$ defining the temperature of 250 magnetic compensation ($T_{comp} \sim 250K$). The horizontal dotted line corresponds to H = 0.55 T which is the maximum 251 magnetic field available on the femtoslicing end-station [37]. The Co₈₀Dy₂₀ alloy layer used for SQUID 252 measurements was deposited simultaneously with the one used for the time-resolved experiments deposited on 253 transparent SiN membrane.





257 Figure 2: Sketch of the experimental conditions to achieve our different relative temperatures $T^* = T_{Curie} - (T_{cryo} - T_{cryo} - T_{cr$ 258 ΔT) = 350K (a), 400K (b) and 540K (c).



Figure 3: Hysteresis loops recorded by monitoring the X-ray transmission at the Dy M_5 absorption edge as a function of the magnetic field. The experimental configurations were P = 0mW and $T_{cryo} = 300K$ (black circles), P= 0mW and $T_{cryo} = 320K$ (red circles) and P = 50mW and $T_{cryo} = 80K$ (black circles).



Figure 4: Transient XMCD at the Co L_3 (black squares) and Dy M_5 (red circles) edges as a function of the pump - probe delay measured at $T^* = 350$ K (a), 400 K (b) and 540 K (c). The solid lines are the fitting functions. The vertical blue dotted lines denote the delay at which the magnetization of the Co sublattices reaches the minimal value.



Figure 5: Characteristic demagnetization times for Co (black squares) and Dy (red circles) sublattices as a function of T*. The error bars on the characteristic demagnetization times is given by the standard deviation from

the fitting function. The error bars on the temperature were experimentally estimated (details in the text).

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Figure 6: Characteristic demagnetization times for Co (filled symbols) and Dy (open symbols) sublattices as a
 function of T*. We report our experimental results (black squares and red circles) superposed to published data
 extracted from element and time-resolved experiments performed on Co₈₀Dy₂₀ (filled and empty blue triangles)
 [27], Co₇₈Dy₂₂ (filled and empty green lozenges) [27] and Co₈₃Dy₁₇ (filled and empty magenta stars) [25].

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- 280

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