## Element- and time-resolved dynamics in rareearth/transition metals alloys

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**Abstract:** Ultrashort laser pulses are used to induce changes of the magnetization in ferrimagnetic  $Co_x Tb_{1-x}$  alloys. Ultrafast magnetization dynamics has been probed by tr-XMCD at the  $CoL_3$  and  $TbM_5$  edges. We demonstrated that demagnetization of the 4f magnetic moment is much faster than in pure Tb when the excited-state-temperature is below the compensation temperature.

## Introduction

Ultrafast magnetization dynamics is an important issue for both fundamental science and for applications in order to optimize spin manipulation on a microscopic level. Application of ultrashort laser pulses allows ultimately the manipulation of the magnetization of magnetic films down to the femtosecond time scale. Therefore it is essential to understand the different fundamental processes taking place during the first hundred femtoseconds. Since the first observation of laser induced spin dynamics [1] the mechanisms of femtosecond demagnetization have been widely debated. The standard technique used in these studies is time-resolved magneto-optic Kerr effect. Nowadays time-resolved X-ray magnetic circular dichroism (TR-XMCD) using synchrotron light with femtosecond time resolution is accessible as a complementary and element-specific tool to study ultrafast magnetization dynamics [2-5]. It is now possible to measure absolute values of the magnetization with a high temporal resolution (130 fs) using the f-slicing source at the Helmholtz-Zentrum Berlin [2-3]. One of the most striking riddle deals with the intrinsic time scale of dynamics, faster for 3d transition metals (TM) (~100fs) than

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for 4f rare earths (RE) (~700 fs) [4]. In RE/TM alloys, the indirect 4f-4f coupling is enhanced due to the presence of 3d electrons in the conduction band. It allows investigating the effect of  $3d_{TM}$ - $5d_{RE}$  exchange coupling. Recently, in ferrimagnetic Fe.<sub>75</sub>Co.<sub>34</sub>Gd<sub>.22</sub> it was shown that Gd shows a long demagnetization time of 430 fs whereas Fe is much faster despite the strong exchange interaction between both elements [5]. The numerical values of the thermalization times have not been explicitly addressed.

In this context, we have used laser pulses to induce changes of the magnetization in ferrimagnetic  $Co_x Tb_{1-x}$  at different working temperatures. The magnetization dynamic has been probed at the  $CoL_3$  and  $TbM_5$  edges [6]. We show that the demagnetization time of the 4f magnetic moment can be reduced to values down to  $\tau = 280$  fs compared to pure Tb ( $\tau$ =740 fs). Our observations also suggest that in case of multisublattice ferrimagnets the 4f demagnetization is faster when the excited-state temperature is below the compensation temperature ( $T_{comp}$ ) whereas it is slower when the excited-state temperature approaches the Curie temperature ( $T_c$ ) [6].

The pump fluences used during our experiment were adjusted to  $12 \text{ mJ/cm}^2$  for Co<sub>0.74</sub>Tb<sub>0.26</sub> and 21 mJ/cm<sup>2</sup> for Co<sub>0.86</sub>Tb<sub>0.14</sub> in order to reach similar amplitudes of demagnetization. The 15 nm Co-RE alloys have been grown on Si<sub>3</sub>N<sub>4</sub> membranes. The films are characterized by T<sub>comp</sub>, where the magnetic moments of the Co and the rare-earth sublattices compensate and by a specific T<sub>C</sub> where the magnetic order is lost. A temperature dependent XMCD study at the CoL<sub>3</sub> and rare-earth M<sub>5</sub> edges has been performed to characterize the different films ( $T_{comp}$  and  $T_C$ ) [6]. The pump-probe experiment was performed at film temperatures close to room temperature or slightly above so that the laser excited state temperature is either in the vicinity of  $T_C$  (Co<sub>0.86</sub>Tb<sub>0.14</sub>) or  $T_{comp}$  (Co<sub>0.74</sub>Tb<sub>0.26</sub>). In Figure 1 a, and b we show the ultrafast dynamics obtained at the Co L<sub>3</sub> and Tb M<sub>5</sub> edges for the Co<sub>0.74</sub>Tb<sub>0.26</sub> alloy. The XMCD amplitudes are normalized to the quantitative static XMCD measurements [6]. A model detailed elsewhere [6] suggests that for localized 4f electrons an increase of the spin relaxation time leads to larger thermalization times in the vicinity of T<sub>c</sub>. This model also explains why only minor changes are observed for 3d electrons in Co. This is confirmed by comparing the ultrafast dynamics at the TbM<sub>5</sub> edge in Co<sub>0.74</sub>Tb<sub>0.26</sub> (~280fs) and Co<sub>0.86</sub>Tb<sub>0.14</sub> (~500fs) where the excited-state temperature is below  $T_{comp}$  resp.  $T_C$  (Figure 1 b and c). Finally, we wish to point out that although the 4f elements studied so far are demagnetization rate limited close to T<sub>C</sub> by the divergence of 4f spin fluctuations, this can be overcome using T<sub>comp</sub> in the ferrimagnetic alloys. In the future, more studies on specific ferrimagnetic materials, consisting of two antiparallelcoupled sublattices, are requested to fully describe this effect.



**Fig. 1. (a, b)** Ultrafast dynamics of Co L<sub>3</sub> (triangles) and Tb M<sub>5</sub> (open circles) edges in Co<sub>0.74</sub>Tb<sub>0.26</sub>. (c) Ultrafast dynamics of Tb M<sub>5</sub> edge (open squares) in Co<sub>0.86</sub>Tb<sub>0.14</sub>.

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## References

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- 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] C. Stamm et al., "Femtosecond modification of electron localization and transfer of angular momentum in nickel" Nature Mater. 6, 740-743 (2007).
- [3] C. Boeglin, E. Beaurepaire, V. Halté, V. Lopez-Flores, C. Stamm, N. Pontius, H. A. Dürr, and J.-Y. Bigot, "Distinguishing the ultrafast dynamics of spin and orbital moments in solids" *Nature* 465, 458-461 (2010).
- [4] M. Wietstruk et al., "Hot-Electron-Driven Enhancement of Spin-Lattice Coupling in Gd and Tb 4f Ferromagnets Observed by Femtosecond X-Ray Magnetic Circular Dichroism" Phys. Rev. Lett. 106, 127401 (2011).
- [5] I. Radu et al., "Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins" *Nature* 472, 205-209 (2011).
- [6] V. López-Flores et al. "Role of critical spin fluctuations in ultrafast demagnetization of transition-metal rare-earth alloys" Phys. Rev. B 87, 214412 (2013).