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Layer-sensitive magneto-optical spectroscopic study of magnetization dynamics in multilayered RE-TM structures

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Using spectroscopical magneto-optical methods, we are able to disentangle the contributions of the different layers in the net magnetization of a GdFeCo/SiN/GdFeCo heterostructure. By static magneto-optical measurements, time-resolved magneto-optical Kerr effect spectroscopy and temperature and laser fluence dependence study of the magnetization dynamics, we study the laser-induced magnetization dynamics and reversal in this multilayer system for each layer separately.

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It is well known, that a femtosecond optical laser pulse alone can manipulate the magnetic order1–3 and even trigger a full magnetization reversal of metallic samples.4 The latter is called all-optical switching (AOS) and was first demonstrated in GdFeCo,4,5 a rare earth–transition metal (RE-TM) ferrimagnetic amorphous alloy, near its magnetization compensation temperature ($T_M$). Being a function of the alloy’s chemical composition, this $T_M$ plays an important role in the magnetization dynamics. It was in fact demonstrated6,7 that both AOS and the ultrafast demagnetization efficiency are strongly dependent on the difference between the initial temperature of the experiment $T$ and the $T_M$. Engineering of new and more sophisticated magnetic structures, for instance, by stacking RE-TM amorphous alloy layers with different RE-concentrations, is likely to adjust the $T_M$ of the whole sample, which can result in the possibility for AOS in a much wider range of temperatures than for a single layer case. Practically, the full understanding of the complex net magnetization dynamics requires to have a direct access to each separate magnetic layer. For layers made from different materials, the element-specific soft X-ray and EUV techniques8–10 have been used. These techniques probe the transitions of strongly localized electrons of which their exchange split energy levels serve as the fingerprint of the element’s magnetic state. For table-top experiments with light in the visible spectral range, this can sometimes also be done.11 However, if all magnetic layers of a chosen structure are made of the same material, measuring the total net magnetization dynamics requires some other layer-sensitive technique.

Electromagnetic wave theory for magnetic layered systems12 developed by an extension of Yeh’s formalism,13 which itself is based on the generalization of Abeles’ and Jones’ matrix methods.14,15 predicts the feasibility to distinguish the magneto-optical (MO) response from various magnetic layers in the multilayered structures. The first experimental demonstration of such a separation was achieved by static magneto-optical Kerr (MOKE) ellipticity measurements of Co/Au multilayers,16 where hysteresis loops for specific Co layers were obtained by varying the illumination photon energy. The MOKE spectra of those materials demonstrated features, which mainly reproduced the spectral dependence of the optical parameters of Au,17 with pronounced maxima of Kerr rotation and a sign change of Kerr ellipticity in the region of the plasma edge energy of gold. However, GdFeCo amorphous alloys have no significant spectral features in the visible range.

In this work, we investigate the relative impact of the individual layers in a GdFeCo/SiN/GdFeCo structure on its total magnetization and magnetization dynamics, by static and time-resolved MOKE spectroscopy. Our experimental results demonstrate that spectroscopy can be used to separate the magnetic contributions from different RE-TM layers of this system. By employing this MOKE spectroscopy tool and conducting a temperature and fluence dependence magnetization dynamics study, we observe the layer-specific laser-induced magnetization reversal in this multilayer structure.

The series of GdFeCo/SiN/GdFeCo films was deposited by magnetron sputtering, with concentrations $x$ and $y$ varied between 22% and 27%. All the samples are transparent in the visible range and demonstrated a perpendicular magnetic anisotropy. In this paper, we discuss the spectroscopic MOKE results of a SiO2/AlTi/SiN/Gd$_{26}$Fe$_{64.7}$Co$_{9.3}$/SiN/Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$/SiN structure, with 10 nm-thick AlTi and magnetic layers and 5 nm-thick dielectric spacers. Employing the MO Faraday effect, we extracted the temperature dependence of the coercive field ($H_c$) from the hysteresis loops (see Fig. 1). The magnetization loops in Fig. 1(a) demonstrate the superposition of the MO responses from two individual magnetic layers and do not originate from the second order effects, as outlined in Ref. 17. The coercive field divergence in Fig. 1(b) indicates the $T_M$. The hysteresis loop shape analysis as a function of temperature reveals the existence of two magnetization compensation temperatures at $T_{M1} \approx 210$ K and $T_{M2} > 315$ K. We explain this observation by the fact that the layers are separated by a 5 nm-thick...
dielectric spacer and, therefore, do not interact with each other. With an external magnetic field applied in the same direction, the relative magnetization direction of Gd for the two layers depends on temperature. Thus, \( M_{Gd26} \) is parallel to \( M_{Gd27} \) for temperatures \( T < T_{M,1} \), while for \( T_{M,1} < T < T_{M,2} \) the antiparallel alignment is present, as shown in Fig. 1(b).

For a static MO characterization, the magnetic field was applied perpendicularly to the sample surface. The multilayer was illuminated by linearly polarized laser light with a wavelength of \( \lambda_{in} \) varied in the spectral range from 420 nm to 750 nm and an angle of incidence \( \alpha_{in} \) varied between 0° and 45° to the sample normal. The results shown in Figs. 2(a) and 2(c) reveal a difference in sensitivity of the MO response to different layers for the experiments performed in Faraday (transmission) and Kerr (reflection) configurations, respectively. Figures 2(a) and 2(b) show the results obtained in the Faraday geometry at \( T = 270 \) K, \( \lambda_{in} = 600 \) nm and normal incidence from the AlTi-side. Note that any variations of either \( \lambda_{in} \) or \( \alpha_{in} \) of the incident light had no visible effect on the hysteresis loop shape. By analysing the shape and conducting the superposition decomposition depicted in Fig. 2(b), we were able to conclude that in the Faraday configuration, where the laser beam is detected after being transmitted through the sample, the two magnetic layers contribute for 49% and 51% to the total magneto-optical signal, respectively. The \( Gd_{27}Fe_{63.9}Co_{9.1} \) layer contribution is seen to be slightly lower compared to the \( Gd_{26}Fe_{64.7}Co_{9.3} \) one, which is expected, due to light absorption in the \( Gd_{26}Fe_{64.7}Co_{9.3} \) layer before the light interacts with \( Gd_{27}Fe_{63.9}Co_{9.1} \). In the Kerr geometry (see Fig. 2(c)), where the light reflected from the sample is detected, the layer sensitivity could be tuned at will by adjusting either \( \lambda_{in} \) or \( \alpha_{in} \). Thus, the MOKE spectroscopy revealed changes in the hysteresis shape obtained at various wavelengths of the laser excitation. By heating the sample optically at negative pump-probe delay (\( t < 0 \)) and looking at the shapes of the magneto-optical hysteresis loops, we were

FIG. 1. Magneto-optical static characterization of the \( Gd_{26}Fe_{64.7}Co_{9.3}/SiN/ Gd_{27}Fe_{63.9}Co_{9.1} \) structure. (a) The hystereses obtained in the Faraday geometry at \( T = 90 \) K, 180 K, 220 K, and 270 K. A superposition of magneto-optical responses from two magnetic layers is detected. (b) Coercive field temperature dependence extracted from the hysteresis loops. Coercive field divergence indicates the magnetization compensation temperatures around \( T_{M,1} = 210 \) K for \( Gd_{26}Fe_{64.7}Co_{9.3} \) layer and above \( T_{M,2} = 315 \) K for \( Gd_{27}Fe_{63.9}Co_{9.1} \) layer. The magnetization direction of Gd for the two GdFeCo layers is parallel for \( T < T_{M,1} \) and antiparallel for \( T_{M,1} < T < T_{M,2} \).

FIG. 2. Magneto-optical static characterization obtained in the Faraday (in transmission) and the Kerr (in reflection) geometries at \( T = 270 \) K. (a) The Faraday characterization reveals a layer sensitivity independence from wavelength or angle of incidence of the laser illumination. (b) A simulation of a superposition decomposition, obtained in the Faraday geometry. (c) Magneto-optical Kerr spectroscopy in the range of \( \lambda_{in} = 420 \) nm–750 nm at \( \alpha_{in} = 45^\circ \) of incident angle. The magneto-optical sensitivity to different layers is strongly dependent on the wavelength of incident light.
able to confirm that the layer sensitivity in Kerr spectroscopy is independent from the temperature.

Due to a difference between the coercive fields of Gd$_{26}$Fe$_{64.7}$Co$_{9.3}$ and Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ at $T = 270 \text{ K}$, we were able to separate the changes in the MO signal from the different layers (see Fig. 3). Consequently, we could determine the spectral regions where the MO response of the whole structure originates from one particular layer only. Our results show that for this particular sample at $\lambda_{in} = 610 \pm 5 \text{ nm}$ and $\sigma_{in} = 45^\circ$ laser light illumination, the total net MO response originates from the Gd$_{26}$Fe$_{64.7}$Co$_{9.3}$ part only. By tuning the wavelength to $\lambda_{in} = 670 \pm 5 \text{ nm}$ at the same angle of incidence, this sensitivity shifts to the Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ layer exclusively. Outside these spectral regions, the hysteresis loop shape obtained in the Kerr geometry is seen as a weighted superposition of hysteresis loops from the two magnetic layers. Note that the MO phase of the signal from different layers depends on the wavelength. Indeed, in the spectral region from 420 nm to 610 nm, the hysteresis loops from the two layers demonstrate opposite signs. For longer wavelengths, the phase of the Kerr signal from the Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ layer crosses zero after which the sign of the hysteresis loops becomes identical. Increasing the wavelength beyond 670 nm, the phase of the Kerr signal from the Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ layer changes and the sign of the hysteresis loops becomes opposite again. Thus, our results reveal the feasibility of layer-selective MO spectroscopy in the RE-TM materials. This opens the possibility to separate the changes in the MO signal from the different layers, which can lead to a better and more accurate understanding of MO studies of complex magnetic heterostructures.

Next, we will demonstrate how this layer-selective spectroscopy can be implemented in a magnetization dynamics study by means of time-resolved MOKE measurements. During the experiment, the multilayer was illuminated by a linearly polarized 60-fs laser excitation with a laser repetition rate of 1 kHz, a central wavelength of $\lambda = 800 \text{ nm}$, a beam-radius of $\sigma = 200 \mu \text{m}$, and a fluence ranging from 1 mJ/cm$^2$ to 10 mJ/cm$^2$. The magnetization state was subsequently probed by a time-delayed single linearly polarized laser pulse ($\tau \approx 60 \text{ fs}$, $\lambda = 410 \text{ nm–}600 \text{ nm}$, $\sigma \approx 80 \mu \text{m}$, and a laser fluence <0.1 mJ/cm$^2$). We note here that the magnetization dynamics study required an experimental setup different from the one used for static characterization. The optical parametric amplifier did not let us use 610 nm and 670 nm light for the direct probing of the magnetization dynamics in different layers. Therefore, we measured the magnetization dynamics at different wavelengths, where the MO superposition from both layers was present, and from those measurements derived the magnetization dynamics for each layer. The external magnetic field of $H_{ext} = 340 \text{ mT}$ was applied perpendicularly to the sample surface. The experiments were conducted at $T$ in the range from 80 K to 310 K.

Figure 4 demonstrates the MOKE spectroscopy results obtained by the femtosecond time-resolved magneto-optical stroboscopic setup at $T = 170 \text{ K}$, $\sigma_{in} = 45^\circ$ and the wavelengths of $\lambda_{in,1} = 560 \text{ nm}$ [Figs. 4(a) and 4(c)] and $\lambda_{in,2} = 420 \text{ nm}$ [Figs. 4(b) and 4(d)]. From the hysteresis loop shapes shown in Figs. 4(a) and 4(b), we were able to determine the contributions from the different magnetic layers to the total net MO response of the whole system. Thus, for $\lambda_{in,1} = 560 \text{ nm}$, the ratio was 60%/40% while for $\lambda_{in,2} = 420 \text{ nm}$ the layers contributed as 23%/77% for Gd$_{26}$Fe$_{64.7}$Co$_{9.3}$ and Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ parts, respectively. The laser fluence dependence of the magnetization dynamics shown in Figs. 4(c) and 4(d) demonstrates the quenching of the magnetization after the laser excitation, followed by a magnetization relaxation. The amount of demagnetization scales linearly with the laser fluence. A comparison of the results shown in Figs. 4(c) and 4(d) reveals the differences in the magnetization dynamics behavior during the first nanosecond after the laser excitation, as well as in the final magnetic state. To decompose the superposition of the MO responses from the two layers, we determined the magnetization dynamics of the Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ layer as

$$\frac{77\% \times \left[M(t)/M_0\right]_{n=1} - 40\% \times \left[M(t)/M_0\right]_{n=2}}{60\% \times 77\% - 23\% \times 40\%}$$

as well as for the Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ layer as

![FIG. 3. The wavelength dependence of the magneto-optical sensitivity to 10 nm-thick GdFeCo layers, separated by a 5 nm-thick SiN spacer, obtained at $T = 270 \text{ K}$. The sample is illuminated at $\sigma_{in} = 45^\circ$. At $\lambda_{in} = 610 \pm 5 \text{ nm}$ and $\lambda_{in} = 670 \pm 5 \text{ nm}$, the magneto-optical response from the whole structure is seen exclusively from the Gd$_{26}$Fe$_{64.7}$Co$_{9.3}$ and Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ layers, respectively.](image)

![FIG. 4. Static and time-resolved magneto-optical Kerr spectroscopy of Gd$_{26}$Fe$_{64.7}$Co$_{9.3}$/SiN/Gd$_{27}$Fe$_{63.9}$Co$_{9.1}$ structure obtained at $T = 170 \text{ K}$. The magnetic state of the sample is probed by $\lambda_{in,1} = 560 \text{ nm}$ [Figures (a) and (c)] and $\lambda_{in,2} = 420 \text{ nm}$ [Figures (b) and (d)]. 60 fs laser pulses. Figures (a) and (b) demonstrate the magneto-optical hysteresis at these wavelengths. Figures (c) and (d) represent the laser fluence dependence of the magnetization dynamics superpositions.](image)
To conclude, in this work, we experimentally demonstrated the feasibility of layer-selective MOKE spectroscopy in the RE-TM materials. By implementing this technique for a time-resolved magneto-optical study, we were able to decompose the net magnetization dynamics of a heterostructure into the magnetization dynamics of each individual layer. Together with the element-specific probing proposed in Ref. 11, this layer-selective spectroscopy technique may encourage table-top magnetization dynamics studies of magnetic multilayers engineered for all-optical switching.

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FIG. 5. The laser fluence dependence of the magnetization dynamics of Gd26Fe64.7Co9.3 and Gd27Fe63.9Co9.1 layers, extracted from the superposition shown in Figures 4(c) and 4(d). The ultrafast laser-induced magnetization reversal is detected for the Gd26Fe64.7Co9.3 layer, while partial demagnetization and field-induced magnetization reversal above $T_{M,2}$ is observed for Gd27Fe63.9Co9.1.

\[ \frac{23\% \times \left[ M(t)/M_0 \right]_{\alpha,1} - 60\% \times \left[ M(t)/M_0 \right]_{\alpha,2}}{23\% \times 40\% - 60\% \times 77\%} \]  

(2)

The decomposed laser fluence dependence for the magnetization dynamics of the Gd26Fe64.7Co9.3 and Gd27Fe63.9Co9.1 layers is shown in Fig. 5. Three regimes of the magnetization dynamics are observed. For the laser excitation fluence of 4 mJ/cm², a partial ultrafast demagnetization is seen for both layers, followed by a magnetization relaxation of the Gd27Fe63.9Co9.1 and field-induced magnetization reversal above $T_{M,1}$ for Gd26Fe64.7Co9.3. Increasing the laser power to 5.6 mJ/cm² results in the observation of field-induced magnetization reversal of Gd26Fe64.7Co9.3 and Gd27Fe63.9Co9.1 above $T_{M,2}$. For the laser pump fluence of 7.4 mJ/cm², AOS is observed for Gd26Fe64.7Co9.3 layer, while Gd27Fe63.9Co9.1 layer demonstrates an increased speed of a field-induced reversal above $T_{M,2}$. The obtained results fit very well with the typical magnetization behavior of a GdFeCo amorphous alloy in the proximity of $T_M$ and at temperatures of the experiment $T \ll T_M$.7