Ultrafast non-local spin dynamics in metallic bi-layers by linear and non-linear magneto-optics

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Abstract We make a step towards the understanding of spin dynamics induced by spin-polarized hot carriers in metals. Exciting the Fe layer of Au/Fe/MgO(001) structures with femtosecond laser pulses, we demonstrate the ultrafast spin transport from Fe into Au using time-resolved MOKE and mSHG for depth-sensitive detection of the transient magnetization.

Ultrafast spin dynamics is the key for development of data storage and spintronics devices. Modern all-optical techniques provide ultimate time resolution of sub-10 fs for the related studies. However, recent findings on laser-induced magnetization dynamics [1] still challenge the understanding of ultrafast magnetism. With this contribution, we make a step towards the understanding of ultrafast spin dynamics in metallic multi-layers, which is (i) spatially non-uniform due to interfaces and strong absorption of the pump pulse and (ii) non-local due to highly mobile hot carriers (HC) that are spin-polarized if excited in a ferromagnet.

The transient magnetization **M** was monitored by the magneto-optical Kerr effect (MOKE) and magneto-induced second harmonic (SH) generation (mSHG) using the 800 nm, 14 fs output of a cavity-dumped Ti:sapphire oscillator. The SH intensity $I^{2\omega}(\mathbf{M},t) \propto |\mathbf{E_{even}}+\mathbf{E_{odd}}|^2$, where t is the pump-probe delay, $E_{even}(t)$ is independent of **M** and $E_{odd}(t) \propto M(t)$. The interface spin dynamics is analyzed by $\Delta_{odd}(t) \approx E_{odd}(t)/E_{odd}(t_0) - 1 \approx M(t)/M_0 - 1$ defined from $I^{2\omega}(\pm M,t)$ and $I^{2\omega}(\pm M_0,t_0)$; t_0 represents the absence of excitation [2]. The bulk **M** is probed by $\Delta_{MR,\,ML} \equiv \theta(t)/\theta_0 - 1 \approx M(t)/M_0 - 1$, where θ is the MOKE rotation (MR) or ellipticity (ME). The results obtained in epitaxial Au/Fe/MgO(001) [3] serving as a model system, show pronounced dependence of Δ_{MR} and Δ_{odd} on the Fe thickness d_{Fe} (Fig.1 a, b). This is explained by an increase of the Fe/Au interface contribution with reducing d_{Fe} due to absorption (at 800 nm the light penetration depth in Fe $\lambda_\omega^{Fe} \approx 20$ nm), which is corroborated by the transient reflectivity and $E_{even}(t)$ (not shown).

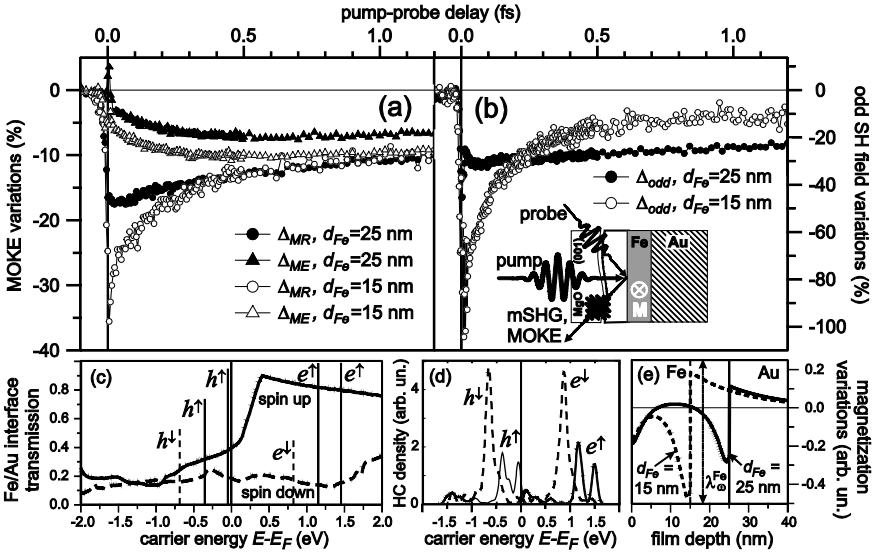

Fig. 1. Pump-induced variations of (a) MOKE rotation and ellipticity, and (b) odd SH field E_{odd} measured in the longitudinal, p-probe and transversal, p-P (inset) geometries, respectively. Experiments were performed for p-polarized pump with the fluence on the order of 1 mJ/cm^2 in Au/Fe/MgO(001) bi-layers with layer thicknesses d_{Au}=60 nm and d_{Fe}=25 and 15 nm, as indicated. (c) Average spin-dependent transmission probability of the Fe/Au interface per available single-electron channel in Fe, calculated from first principles with the Smeagol code [4]. (d) Calculated density of hot carriers [3] excited in Fe by 1.5 eV photons; peak positions are marked in Panel (c) by vertical lines. (e) Variations of the magnetization ΔM after the excitation for 15 and 25 nm Fe films in contact with bulk Au; the model accounts only for the ballistic transport; calculations are based on the data from Panels (c) and (d), and Table 1.

The complete description of the observed transients requires an advanced model like that one developed for the superdiffusive HC transport in Ni on Al [5]. Here we focus on the fastest (within our 20 fs resolution) timescale of the Δ_{MR} and Δ_{odd} break-down (Fig.1 a, b) and model the ballistic HC transport. The related HC parameters are summarized in Table 1 and Fig. 1 c, d. Owing to the largest λ_{Fe}, smallest λ_{Au}, and largest transmission of Fe/Au interface (Fig. 1 c), majority electrons $e^{\uparrow}$ dominate the ballistic spin transport. After the ballistic regime has passed at delays t >10 fs, i.e. $t > \tau_{Fe}$ (see Table 1), M is reduced in Fe and increased in Au near the Fe/Au interface (Fig. 1 e). To explain the different behavior of $\Delta_{MR}(t)$ and $\Delta_{ME}(t)$ (Fig. 1 a) with this model, we make following assumptions: (i) Owing to the large spin-orbit coupling, the transiently magnetized Au provides a MOKE response. (ii) Since at 1.5 eV photon energy Au has high reflectivity but low absorption ($|\mathrm{Re}\,\varepsilon| >> |\mathrm{Im}\,\varepsilon|$), it contributes much stronger to MR (defined by Re ε) than to ME (defined by Im ε). (iii) The Au contribution to MR has opposite sign with respect to the Fe one, which can be explained by a non-resonant character of the Au optical response (in contrast to the resonant case of Fe). This together with $\Delta M(t>0)$ (Fig. 1 e) explains the strong break-down of Δ_{MR}, which increases with

reducing d_{Fe}. Due to similar reasons the reduction of M at the Fe side of the Fe/Au interface and the build-up of M at the Au side after the excitation (Fig. 1 e) leads to strong break-down of Δ_{odd} (Fig. 1 b), which increases with reducing d_{Fe}. This effect however is more pronounced than the break-down of Δ_{MR} due to the interface sensitivity of mSHG. The following dynamics is driven by the superdiffusive HC transport [5] and broadens the ΔM distribution across the Fe film and the Fe/Au interface, which leads to the recovery of Δ_{odd} and Δ_{MR} and further reduction of Δ_{ME}.

In conclusion, we have demonstrated the ultrafast spin transport across the (buried) Fe/Au interface and shown that the combination of time-resolved MOKE and mSHG is a powerful tool to study the non-local spin dynamics in metals.

Table 1. Calculated [3] ballistic velocities of the excited HC in Fe and Au, v_{Fe} and v_{Au}, lifetimes τ_{Fe} and τ_{Au} estimated from Refs. [6], and resulting ballistic propagation lengths λ_{Fe} and λ_{Au}.

HC	v_{Fe} (nm/fs)	τ_{Fe} (fs)	λ_{Fe} (nm)	v_{Au} (nm/fs)	τ_{Au} (fs)	λ_{Au} (nm)
$e^{\uparrow}$	0.43	8	3.4	0.95	15	14
$h^{\uparrow}$	0.08	~8	~0.6	0.77	200	154
$e^{\downarrow}$	0.30	2	0.6	1.17	40	47
$h^{\downarrow}$	0.21	~2	~0.4	0.94	80	75

Acknowledgments The authors thank A.I. Lichtenstein and P.M. Oppeneer for fruitful discussions. Funding by the DFG (ME 3570/1, Sfb 616) and by the EU 7-th framework program (CRONOS) is gratefully acknowledged.

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