Ultrafast laser-excited spin transport in Au/Fe/MgO(001): Relevance of the Fe layer thickness

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Abstract Propagation dynamics of spin-dependent optical excitations is investigated by back-pump front-probe experiments in Au/Fe/MgO(001). We observe a decrease for all pump-probe signals detected at the Au surface, if the Fe thickness in increased. Relaxation processes within Fe limit the emission region of ballistic spins at the Fe/Au interface to ~1 nm.

Recently, we have established magneto-optical femtosecond back-pump front-probe experiments [1] in order to provide insight into spin-dependent (i) transport contributions in ultrafast magnetization dynamics and (ii) non-equilibrium transport in metallic films in general.

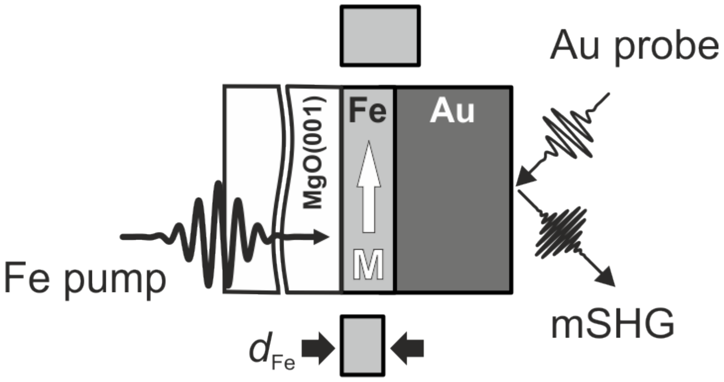

Fig. 1. Scheme of back-pump front-probe experiment. The fs pump pulse at 800 nm is absorbed in the metallic bilayer after transmission through MgO(001) depending on the investigated Fe layer thickness d_{Fe}, here 2, 11, and 17 nm. The dynamics of excitations propagating through the bilayer stack to the Au surface is probed by magneto-induced second harmonic generation (mSHG) as a function of time delay between pump and probe pulses.

As depicted in Fig. 1, the investigated Fe/Au bilayer stack, which is grown epitaxially on MgO(001) [1], is photo-excited by femtosecond laser pump pulses of 14 fs pulse duration and at 800 nm wave length, which are transmitted through the transparent substrate. The probe is sent to the Au surface in order to analyze the propagation dynamics of the excitations through the bilayer stack - similar to a time-of-flight experiment. Here, we use second harmonic generation to detect the excitations arriving at the Au surface for different Fe thickness.

To become highly sensitive to spin-dependent excitations and their propagation dynamics we detect the second harmonic (SH) intensity for opposite Fe magnetization directions. The SH intensity $I_{2\omega}(\mathbf{M},t) \propto |\mathbf{E}_{\mathbf{even}}+\mathbf{E}_{\mathbf{odd}}|^2$, $E_{\mathrm{even}}(t)$ is independent of $\mathbf{M}$ and $E_{\mathrm{odd}}(t) \propto M(t)$. Below, we discuss the SH magnetic contrast $\rho(t)$ and the pump-induced change Δ_{even} of E_{even}; E^0_{even} is the value before optical excitation.

$$\rho(t) = \frac{I^{\uparrow}_{2\omega}(t) - I^{\downarrow}_{2\omega}(t)}{I^{\uparrow}_{2\omega}(t) + I^{\downarrow}_{2\omega}(t)} \propto M(t) \quad ; \quad \Delta_{\mathrm{even}}(t) = \frac{E_{\mathrm{even}}(t) - E^0_{\mathrm{even}}}{E^0_{\mathrm{even}}} .$$

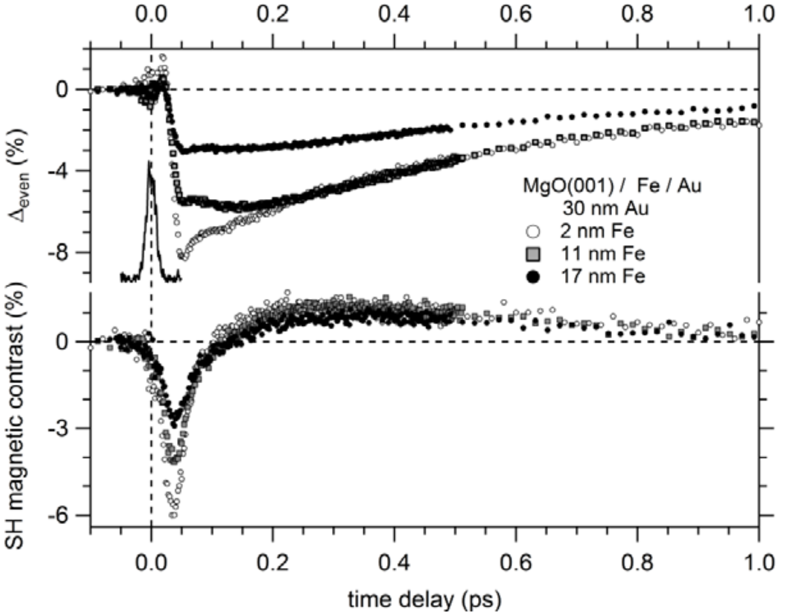

Fig. 2. Results of back-pump front-probe experiments employing SH generation for detecting the propagation of excitations. The top panel shows the relative pump-induced change in the even SH field, the lower panel depicts the relative change of the magnetic contrast $\rho(t)$ for three Fe layer thickness at a fixed Au layer thickness of 30 nm. The solid line in the top panel represents the second harmonic cross correlation which indicates overlap of pump and probe pulses.

Fig. 2 shows the experimental results for three investigated layer stacks of 2, 11, and 17 nm Fe thickness combined with a constant Au thickness of 30 nm. The top panel depicts $\Delta_{\mathrm{even}}(t)$ as a function of pump-probe delay (symbols) and the SH cross correlation of pump and probe pulses (solid line), which indicates time zero. The data exhibit a pronounced reduction at 40 fs delay and a recovery of the initial value within few ps. The variation with the Fe layer thickness d_{Fe} is characterized by a weaker change for larger d_{Fe}.

The delay of the onset of the change originates from propagation of the fastest excitation through the Au layer [1]. In comparison to a variation of the Au layer thickness reported in [1] the variation in d_{Fe} leaves the delay of initial drop fixed. We therefore conclude that the propagation through Au determines the 40 fs delay of initial drop.

In the magnetic contrast we identify a negative and a positive contribution to $\rho(t)$, which were reported to be dominated by ballistic and diffusive carrier propagation, respectively [1]. The isosbestic point near 90 fs close to zero magnetic contrast indicates that the two probed characteristic excitations can be well separated. The minimum value at 40 fs is more pronounced for smaller d_{Fe}. Although weaker, a comparable dependence is identified for the positive change in $\rho(t)$. Both signal contributions imply therefore a larger probability to excite the spin excitations probed at the Au surface for thinner Fe films. The short spin-dependent mean free path (<3 nm) and lifetime (<10 fs) in Fe [2] provides a strong argument that for $d_{Fe}>3$ nm considerable relaxation occurs in the Fe layer, before the excitation can be injected into Au. In other words, the injection of ballistic spin currents into Au is limited to a thin Fe region at the Fe/Au interface. Due to the dominant contribution of injected majority holes [1] with a ballistic mean free path <1 nm which we estimated from electronic lifetimes [2] the active injection region is constricted to a Fe slice of 1 nm thickness at the interface. Thus, the observed effects weaken for larger d_{Fe} since less energy is deposited in the interface region.

The time-dependence of Δ_{even} is regarded as the direct consequence of the carrier redistribution among Fe and Au. The weaker Δ_{even} for larger d_{Fe} is therefore explained by processes in the Fe layer, rather than by light absorption in Au, although the latter occurs for small enough d_{Fe}.

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