Modification of magnetization orientation in Pt/Co/Pt ultrathin films by femtosecond laser pulses

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Abstract. Ultrathin Pt/Co/Pt films were irradiated with femtosecond laser pulses of sufficiently high energy to create irreversible structural and in consequence magnetic modifications. The effect was studied as a function of an initial structure of the Co film, determined by different temperatures during preparation of the Pt buffer layer, and a Co film thickness, which determines the initial out-of-plane or in-plane magnetization orientation. A creation of many magnetic phases is shown. The out-of-plane magnetization state is found to be created for significantly broader fluence range for the Co film deposited on the Pt buffer layer grown at 750°C in comparison to that grown at room temperature. Reversible magnetization dynamics was also studied in the low fluence regime, using a pump-probe setup. The results were discussed using simulations of temperature dynamics.

Laser annealing is an effective method of tailoring magnetic properties of ultrathin magnetic films. Particularly it enables modification of magnetic anisotropy in Pt/Co/Pt [1, 2]. In this work we show the influence of the initial structure of the Co film, determined by the temperature during the growth of the Pt buffer layer, on an appearance of the laser-induced magnetic phases. Laser pulse induced magnetization dynamics was also studied with a support of modeling of pulse-driven temperature changes.

The samples Al₂O₃(001)/Pt(5nm)/Co(d_Co)/Pt(5nm) were produced by the MBE technique. The Pt buffers were grown at room temperature (RT) and at 750°C (high temperature, HT). The other films of the stacks were deposited at RT. The discussed Co thickness d_Co=1.5 and 3.5 nm corresponds to the out-of-plane and in-plane magnetization orientation, respectively. All four combinations of the growth temperature and d_Co were used. Laser annealing was performed using a Ti:sapphire oscillator with a regenerative amplifier. Single laser pulses of duration of 40 fs and wavelength of 800 nm were released manually and focused onto the sample by a lens. Arrays of spots were created at each sample, with pulse energies between 27 and 141 μJ. The laser-annealed samples were studied using a magneto-optical Kerr microscope in polar (PMOKE) configuration. The optical and PMOKE remanence images of the spots revealed a complicated structure of ring-shaped areas of the modified magnetic and optical properties, similar to ones reported before [1, 2]. Similar as before, we assume that any laser-induced irreversible modification depends on the local pulse fluence only. Using a typical analysis method [3] we obtained the fluence values for all boundaries between the regions observed within the spots, as presented in Fig. 1.
Figure 1. Laser-induced magnetization direction, as a function of the pulse fluence, for two values of $d_{Co}$ and two types of the Pt buffer layer. At high fluences the sample is ablated.

Fluences up to about 20 mJ/cm$^2$ do not affect the samples, thus the magnetization direction is unchanged. Above 20 mJ/cm$^2$ a modification starts to occur, and the out-of-plane magnetization state appears also for thicker Co, while for the thinner Co the magnetooptical signal is enhanced (brighter color in Fig. 1). Comparing RT and HT samples, a huge difference can be found in the fluence range responsible for the out-of-plane state. An ablation threshold is much higher for the HT samples. A partial ablation occurs for the thicker Co, but the out-of-plane state with a square hysteresis loop is still present there.

An explanation of some RT/HT differences may be provided by the XRD studies of the RT and HT buffers. The latter one shows a higher crystalline quality and much smoother surface. This means a better growth of following layers. Lower number of defects results in more tight atomic bonds in a crystal, what makes it more difficult to be destroyed e.g. by thermal motion.

To have a clearer picture of the laser annealing process, a temperature dynamics in the pulse laser-heated sample was modeled. A two-temperature model (2TM), involving two interacting baths of electrons and a lattice, was used [4]. Additionally, phase transitions in contributing metals were involved, using standard enthalpy functions, and the sapphire substrate was included, as a heat sink. The bulk thermodynamic parameters of the materials were used in the model, what may cause some errors, however the real “ultrathin” values, which may differ due to e.g. reduced mean free path of the carriers, are in principle unknown. As the model assumes an ideal Pt/Co/Pt structure, it does not distinguish the RT and HT cases. The calculations are demonstrated in Fig. 2(a) for $d_{Co}=1.5$ nm and for the fluence of 30 and 90 mJ/cm$^2$. The electrons absorb the energy from the laser pulse, reaching extreme temperatures, and next they thermalize with the lattice. Structural modifications depend on the increase of the lattice temperature. A cooling rate, determined by a heat transfer to the substrate is also important, as it defines how long the sample stays hot and an effective mixing can take place. According to the results of the calculations, after a 30 mJ/cm$^2$ pulse the lattice temperature is still below the melting point, however atoms can mix intensively [5]. On the contrary, a 90 mJ/cm$^2$ pulse is capable to melt Pt and even boil Co. The calculations for $d_{Co}=3.5$ nm look very similar, however the reached temperatures are a few percent lower, as given energy is distributed over thicker material. In contrast to the qualitative simulation, a real process of melting and boiling within a few ps looks like an explosion, so one can expect ablation, what corresponds well to the experiment.

To cast more light onto the process of laser annealing, the magnetization dynamics was measured using a pump-probe experimental setup. The sample of $d_{Co}=1.5$ nm on the RT Pt buffer was excited by the pump beam of a wavelength of 488nm (from optical parametric amplifier) with a repetition rate of 1 kHz. The magnetization, preferably aligned out-of-plane, was saturated with a perpendicular external magnetic field $\pm 1.6$ kOe. The second less intense beam of a wavelength of 800nm was used to probe the magnetization state in the sample, using a Wollaston prism and a double balanced detector. In Fig. 2(b) the measured
magnetization dynamics is presented for the pulse fluence of 1.8 and 7.9 mJ/cm², which is far below any threshold of single-pulse modification, presented in Fig. 1. An ultrafast change of the magnetic signal is followed by a slow relaxation. The amplitude is fluence-dependent and the direction of changes was found to depend on a direction of the applied magnetic field, what allows us to identify this process as a thermal-origin laser-induced reduction of magnetization (demagnetization) [6].

Experimental value of the long-term decay time is found to be about 300 ps and it corresponds well to the value derived from the simulated curves of the temperature dynamics for the respective fluences, where it is related to a heat transfer into the substrate. With the 7.9 mJ/cm² curve we noticed, that repeating the measurement many times (on a time scale of minutes) the amplitude of demagnetization becomes gradually reduced. This non-repeatability suggests an onset of irreversible modifications in the sample, whose thresholds in the multi-pulse irradiation regime can be significantly lower as compared to the single-pulse case [2]. The results of the simulated temperature dynamics support this suggestion, as the lattice temperature for the 7.9 mJ/cm² pulse reaches about 700 K, which temperature in the classic annealing of Pt/Co systems is reported to bring intermixing of atoms [5]. Obviously, during several tens of picoseconds under 700 K some minor sample variations may happen only, however after thousands of pulses accumulating modifications may finally become significant.

Modifications of magnetization orientation in Pt/Co/Pt ultrathin films by fs laser pulses were found to be strongly dependent on buffer creation conditions. Some results are consistent with the temperature dynamics simulations, however deeper understanding of these effects requires more detailed studies.

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