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Ultrafast Magnetization Dynamics in Epitaxial Ni-Mn-Sn Heusler Alloy Film

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The work is devoted to studies of ultrafast magnetization dynamics induced by femtosecond laser pulses in ferromagnetic Ni-Mn-Sn shape memory Heusler alloy. We studied epitaxial thin $Ni_{54.3}Mn_{31.9}Sn_{13.8}$ film deposited on (001) MgO substrate. Spin precession in an external magnetic field was triggered and detected by the time-resolved magneto-optical Kerr effect (TRMOKE) using pump-probe technique in dual color scheme experiment. Measurements were performed as a function of magnetic field H and pulse power P. The measured TRMOKE signal is composed of oscillatory and background components, both decaying exponentially in a nanosecond time scale. The precession frequency was determined and found to be varying in the range of 1-10 GHz with H up to 3 kOe and decreasing linearly with P. The dependence of Gilbert damping parameter α on H was determined and discussed.

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1. Introduction

Rapid development of femtosecond laser techniques gives an impact in revealing a number of processes occurring from femto- (fs) to picosecond (ps) timescales. It has become possible to study spin dynamics in magnetic materials giving the opportunity for ultrafast information processing. The underlying subtle mechanisms of ultrafast demagnetization phenomenon are still under study [1, 2]. The ultrafast spin-flip scattering, of different type, and/or spin-orbit interaction, plays an important role in the ultrafast demagnetization process [3]. Characteristic timescales of optically excited demagnetization, measured with time-resolved magneto-optical Kerr effect (TR-MOKE), have been reported as of the order of hundreds of fs for 3d- and tens of ps for 4f-ferromagnets [4]. However, in the case of some half-metallic compounds [5, 6] this timescale could extend to hundreds of picoseconds due to temperature transfer channel blocked for electron-spin interaction after laser pulse excitation [7]. This timescale increase is directly related to the phenomenological Gilbert damping coefficient (α) describing energy dissipation in the magnetic system. One of the method allowing for α estimation is ferromagnetic resonance (FMR). The TR-MOKE technique is important to obtain information about frequency (f) and relaxation time (τ) from optically excited magnetization precession. This method was applied in investigation of a number of Heusler alloys [6, 8–17]. In this work we present the results of experimental studies of shape memory $Ni_{54.3}Mn_{31.9}Sn_{13.8}$ epitaxial Heusler alloy film, carrying out the TR-MOKE measurements as a function of applied magnetic field and laser pulse power. Numerical fittings of experimental dependences have been performed and τ, f, and α parameters determined and discussed.

2. Experimental details

The TR-MOKE rotation signal was measured at room temperature for epitaxial alloy film of $Ni_{54.3}Mn_{31.9}Sn_{13.8}$ composition. The film of 100 nm thickness was deposited at 500°C on single crystalline MgO(001) substrate by magnetron sputtering method. At measuring temperature the film remains in austenite phase. The TR-MOKE measurements were performed using 10 kHz repetition rate Ti:sapphire regenerative amplifier laser system, generating $\sim$ 35 fs pulses at 800 nm wavelength. To measure magnetization dynamics, dual color pump-probe technique was used. The pump pulse excites the sample at fundamental wavelength while the time-delayed probe pulse was frequency doubled and detected at 400 nm. The pump and probe beams of s-polarization were directed at $\sim 10°$ and $\sim 40°$ with respect to the sample surface normal. To avoid inhomogeneity in excited material area the pump beam was focused into ~ 200 μm while the probe spot diameter was roughly 4× smaller. The Kerr rotation of the probe beam was synchronously detected with balanced optical bridge. The TR-MOKE effect was measured as a function of external magnetic field amplitude H and pump beam power P. In the first scenario H was applied at $\sim 45°$ with respect to the sample plane in the range up to 2.4 kOe, while in the following one H was fixed at 1.5 kOe and P was varied in the range of 0.15−2.0 mW. In all performed measurements probe beam was fixed at 0.15 mW. Other details concerning the experimental setup are similar as in Ref. [18]. Static

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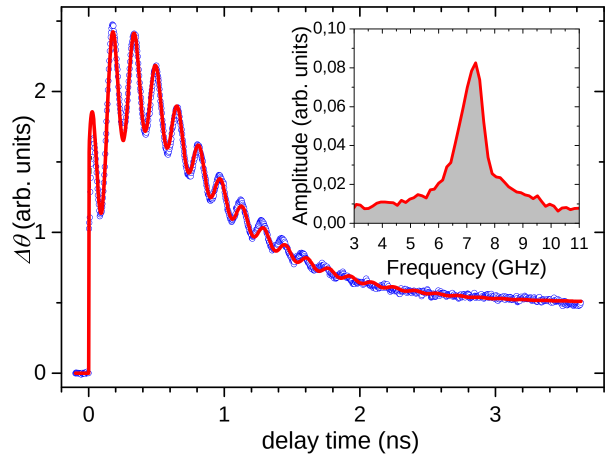

Fig. 1. Typical TR-MOKE signal for $Ni_{54.3}Mn_{31.9}Sn_{13.8}$ Heusler alloy film measured for magnetic field of H = 1.5 kOe at pump beam power of P = 0.8 mW. In the inset, Fourier transform of the oscillatory part of the signal is shown.

magnetic properties of this sample, studies with MOKE, were included and described in detail in Ref. [19].

3. Results and discussion

In Fig. 1 typical dependence of Kerr rotation $\Delta\theta(t)$ induced by pump pulses as a function of time t delayed probe pulses is shown. A characteristic feature of measured dependences is two component processes described by $\Delta\theta(t) = \Delta\theta_{\rm osc}(t) + \Delta\theta_{\rm nosc}(t)$. The oscillatory part $\Delta\theta_{\rm osc}(t)$ is related to periodically oscillating $\Delta\theta$ as a function of t with decaying amplitude. These oscillations have a large background $\Delta\theta_{\rm nosc}(t)$ rapidly increasing at t=0 and then monotonically decaying. Mutual relation between amplitude of oscillations and background components depends strongly on both H and P. To simplify further analysis, we will consider the oscillatory part only of measured dependences. For this purpose $\Delta\theta_{\rm nosc}(t)$ has been approximated with function: $\Delta\theta^{\rm theor}_{\rm nosc}(t) = \mathrm{erf}(2\ln(2)\frac{t-t_0}{\tau_d})[(q_1-q_2)\exp(-\frac{t}{\tau_e})+q_2]$ with the set of fitting parameters [16].

The experimental oscillatory parts $\Delta\theta_{\rm osc}(t) = \Delta\theta_{(}t) - \Delta\theta^{\rm theor}_{\rm nosc}(t)$ were determined for all measured dependences. In the inset of Fig. 1, the fast Fourier transform (FFT) of $\Delta\theta_{\rm osc}(t)$ component is presented. Figure 2 shows selected dependences of $\Delta\theta_{\rm osc}(t)$ for increasing P in the range of 0.15−1.4 mW at fixed amplitude of H = 1.5 kOe. From the dependences presented in Fig. 2 it follows that with increase of P (i) the amplitude of oscillations increases monotonically and tends to saturation for maximum values of P used (see Fig. 3a), (ii) the magnetization precession relaxation time decreases (see Fig. 3e), and (iii) the frequency of oscillations decreases as well (see Fig. 3c). In Fig. 2, theoretical dependences of $\Delta\theta_{\rm osc}(t)$ for selected cases are shown with solid lines which were fitted according to the formula [18]:

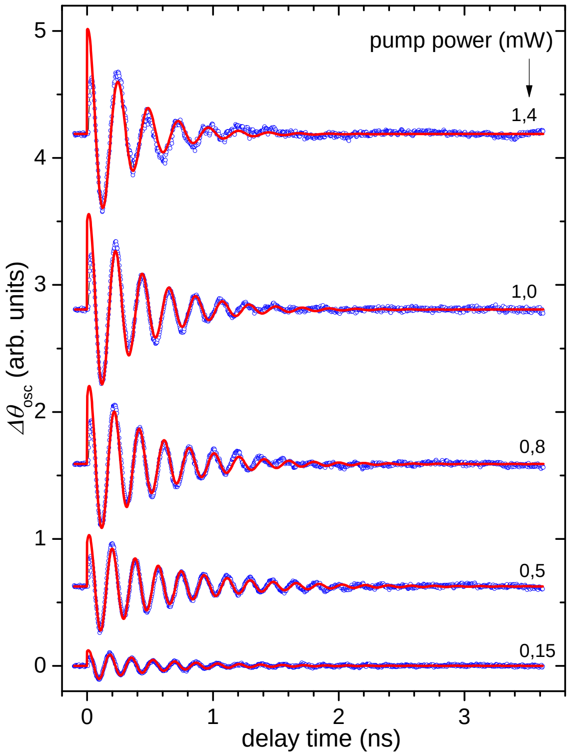

Fig. 2. Oscillatory part of TR-MOKE signal for $Ni_{54.3}Mn_{31.9}Sn_{13.8}$ Heusler alloy film measured for various pump beam powers P at magnetic field of H = 1.5 kOe.

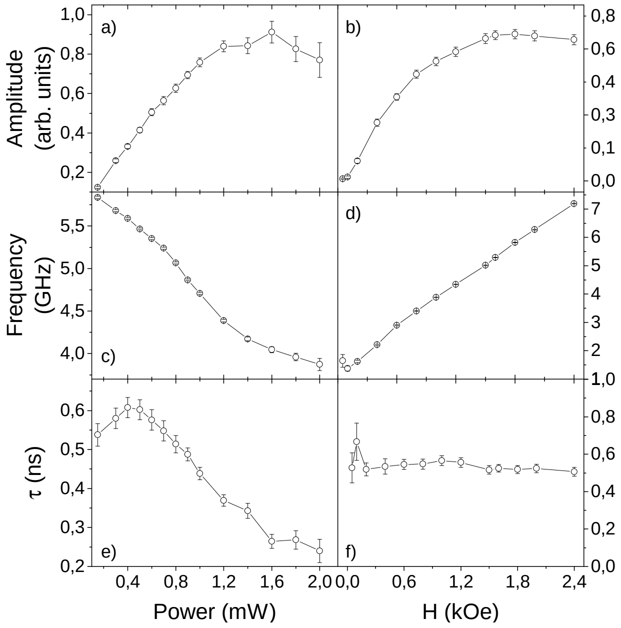

Fig. 3. Determined a,b) amplitudes, c,d) frequencies, e,f) relaxation times of TR-MOKE signal measured as a function of pump beam power P at fixed magnetic field H = 1.5 kOe and magnetic field H at fixed P = 0.8 mW.

$\Delta\theta_{\mathrm{osc}}(t) = A\sin[2\pi(ft+\varphi)]\exp(-t/\tau)$, where φ is oscillations phase. As it can be seen, theoretical curves are in good agreement with the experiment.

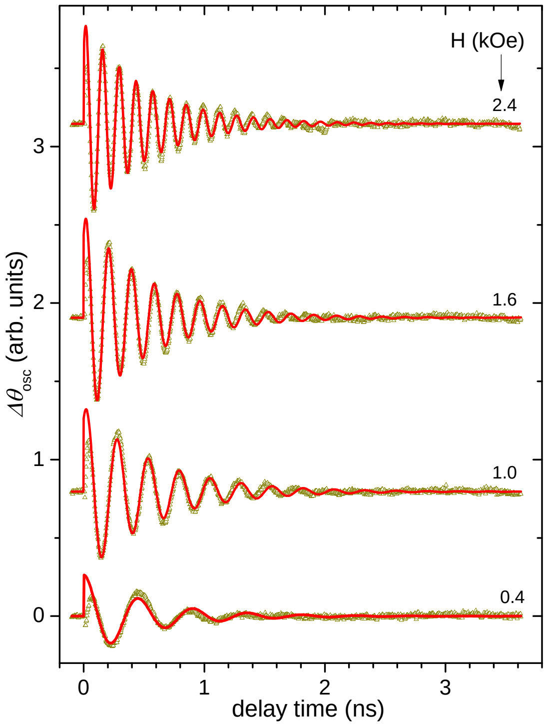

Fig. 4. Oscillatory part of TR-MOKE signal for $\mathrm{Ni_{54.3}Mn_{31.9}Sn_{13.8}}$ Heusler alloy film measured for various magnetic field amplitudes at $P = 0.8$ mW pump beam power.

The $\Delta\theta_{\mathrm{osc}}(t)$ dependences for selected values of H at the fixed pump value of $P = 0.8$ mW are shown in Fig. 4. It can be seen in Fig. 4 that with increase of the H value (i) the amplitude and frequency f of the oscillations increases (see Fig. 3b and 3d, respectively), however (ii) the relaxation time τ does not change noticeably with H (see Fig. 3f). In the following, the oscillation frequency, as a function of the field H and power P, will be considered in detail. The $f(H)$ dependence at fixed power $P = 0.8$ mW is shown in Fig. 5a. The $f(H)$ function is nearly linear and is well described by the Kittel formula $f = \frac{\gamma}{2\pi}\sqrt{H_x(H_x + H_{\mathrm{keff}})}$, where H_x is the in-plane component of the external magnetic field, γ is the gyromagnetic ratio of the electron, $H_{\mathrm{keff}} = 4\pi M_{\mathrm{s}} - H_{\mathrm{ku}}$ is the effective field of magnetic anisotropy, composed of shape anisotropy and uniaxial anisotropy H_{ku}. From the fit of the theoretical function to the experimental $f(H)$ dependence, the value of $H_{\mathrm{keff}} = 1.8 \pm 0.3$ kOe was determined.

Figure 5b shows the dependence of $f(P)$ at a fixed value of field $H = 1.5$ kOe. The three ranges of $f(P)$ linear dependences: 0.15−0.7 mW, 0.7−1.3 mW and 1.3−2.0 mW, can be distinguished. For the second range, changes of $f(P)$ are more pronounced than for the first and last one. The reduction of the oscillation frequency with increase of pumping energy can be associated with a significant disturbance of the electronic structure of the system which affect temporal values of the effective field of magnetic anisotropy [13].

An important parameter characterizing the examined epitaxial layer of the Heusler alloy is the α Gilbert damping parameter. For the exact determination and analysis dependences of α parameter as a function of H and P, the fitting procedure to the measured experimental data of $f(H)$ and $\tau(H)$ should be applied. For this purpose solving equation system, derived from LLG equation, should be performed [20]. For the α parameter estimation, the approximate formula $\alpha = 2[\gamma\tau(2H_x + H_{\mathrm{keff}})]^{-1}$ has been used in this work. Applying the average value of $\tau = 0.54 \pm 0.04$ ns, determined from the experimental data, we obtained that α varies from 0.11 to 0.04 for H fields in the range 0.15−2.4 kOe.

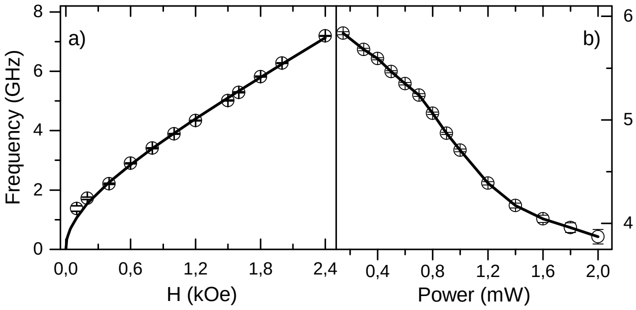

Fig. 5. Magnetization precession frequency for the $\mathrm{Ni_{54.3}Mn_{31.9}Sn_{13.8}}$ Heusler alloy film as a function of: (a) magnetic field amplitude at pump beam power $P = 0.8$ mW and (b) pump beam power P at magnetic field $H = 1.5$ kOe. The solid line in (a) is fitted theoretical dependence described by the Kittel formula (see text).

It is interesting to compare the values of the α parameters determined in the TR-MOKE and FMR experiments. For the stoichiometric $\mathrm{Ni_{50}Mn_{25}Sn_{25}}$ Heusler alloy film the α parameter determined in the FMR experiment is $\alpha = 0.009$ [21]. This is an order of magnitude lower value than the values determined above in the TR-MOKE experiment for the off-stoichiometric $\mathrm{Ni_{54.3}Mn_{31.9}Sn_{13.8}}$ film. It should be emphasized that the values of α parameters determined in the TR-MOKE measurements depend not only on the H field, but also on the P pulse power. A more detailed analysis of the α damping factor as well as the background component $\Delta\theta_{\mathrm{nosc}}(t)$ will be the subject of further study.

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