Ultrafast Demagnetization for Ni$_{80}$Fe$_{20}$ and Half-metallic Co$_2$MnSi Heusler Alloy Films

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Abstract. We investigated ultrafast demagnetization for NM/Ni$_{80}$Fe$_{20}$ (Py)/NM (NM=Ta,Pt) and epitaxial half-metallic Co$_2$MnSi (CMS) films using an all-optical pump-probe technique to clarify the correlation between demagnetization time $\tau_M$ and magnetic damping constant $\alpha$ or spin polarization. The signal from the all-optical time-resolved magneto-optical Kerr effect exhibited rapid decrease in the sub-ps time regime and damped oscillations for these films. Values of $\tau_M$ and $\alpha$ were evaluated using the three-temperature model and the Landau-Lifshitz-Gilbert equation. The $\alpha$ values for the NM/Py/NM films depended on both the Py thickness and NM materials while $\tau_M$ was almost constant. The $\tau_M$ values for the epitaxial CMS films were almost independent of L2$_1$-ordering and a little shorter than those for NM/Py/NM films.

1. Introduction
When conventional ferromagnetic films are irradiated with an intense femtosecond light pulse, spontaneous magnetization decreases within times of sub-ps order. This effect is called ultrafast demagnetization, which can be observed with all-optical pump-probe time resolved Kerr effect (TRMOKE) measurements [1]. According to the three-temperature model, the speed of demagnetization is characterized by the demagnetization time $\tau_M$. This time constant is approximately the same as the electron-spin relaxation time $\tau_{e-s}$, namely the time constant for the energy relaxation process between conduction electron and spin system. The mechanism behind this process, however, is still an open question. Quite recently, Muller et al have verified experimentally the correlation between $\tau_M$ and spin polarization $P$ at the Fermi level in various ferromagnets including certain oxides and Heusler half-metals, and have suggested the relation of $\tau_M/\alpha^{-1}$ [2]. Such a relationship is naturally suggestive if demagnetization originates with spin-flip scattering of spin polarized band electrons via spin-orbit interaction. The density of states at the Fermi level for the minority spin band is negligibly small for type-I half-metals, so that spin-flip scattering is forbidden and $\tau_M$ is extended. Such a trend has been actually observed in the case of some oxide half-metal films, but not in polycrystalline Co$_2$MnSi Heusler alloy (CMS) films even though the CMS alloy is also a type-I half-metal.

From a different point of view, $\tau_M$ is also related to the macroscopic spin relaxation which is characterized by the magnetic damping constant $\alpha$. According to some quantum mechanical theories, one can expect an inverse relationship $\tau_M \propto \alpha^{-1}$ [3, 4, 5, 6]. This has been examined
in Ni$_80$Fe$_{20}$ (Py) films with the addition of Dy or Pd as a third element, although experimental results have shown too weak a correlation to confirm this relationship [7].

Both the spin polarization and magnetic damping constant are the most important factors governing practical applications using spin transfer torque and tunnel magnetoresistance (TMR), and they are necessary in gaining a deeper understanding of the correlations between $P$, $\alpha$ and $\tau_M$. So motivated, we investigated ultrafast demagnetization in normal-metal(NM)/Py/NM (NM=Ta, Pt) and epitaxial CMS films. The NM/Py/NM films are good candidates to investigate $\alpha$-$\tau_M$ relationship because $\alpha$ values can be enhanced arbitrarily by designing the stacking structure [11]. It is likely that $\tau_M$ can vary with $\alpha$ in NM/Py/NM films even if the $\alpha$ enhancement is due to interface effects such as spin pumping, alloying and electronic hybridization. Our epitaxial CMS films exhibit large TMR ratios and $P$ is around 90% evaluated from these TMR ratios [9], in which we expect the $\tau_M$ value to be much larger than that for Py.

2. Experimental Methods

Ta(5 nm)/Py(20 nm)/Ta(5 nm), Pt(5 nm)/Py(10 nm)/Pt(2 nm), and Pt(5 nm)/Py(2 nm)/Pt(5 nm) films were deposited on a glass substrate using magnetron sputtering. The 30 nm thick epitaxial CMS films were grown on Cr-buffered single crystal (001) MgO substrate using UHV-magnetron sputtering and were annealed in vacuum. We prepared CMS films annealed at 300, 450 and 500$^\circ$C. Structural analyses were performed by X-ray diffractometer and pole figure measurements showed that the epitaxial relationship was MgO(001)<100>/CMS(001)<110> for all CMS films. Magnetization was measured by superconducting quantum interference device. TRMOKE measurements were performed at room temperature using the standard optical set-up with a Ti:sapphire laser and a regenerative amplifier. The pump fluence was approximately 3.5 mJ/cm$^2$ while the probe fluence was about ten times smaller. The laser light wavelength was 800 nm and pulse width was ~100 fs. The maximum applied magnetic field was 4 kOe and the field direction was changeable from out-of-plane to in-plane of a film. Details have already been described elsewhere in the literature [8, 9].

3. Experimental Results and Discussion

Fig. 1(a) shows an example of TRMOKE data for NM/Py/NM films measured under an applied magnetic field of 4 kOe at $\theta_H=23^\circ$, where $\theta_H$ is defined as the angle between the magnetic field direction and the film normal. The rapid variation of the TRMOKE signal in the ps time regime corresponds to ultrafast demagnetization and is followed by an oscillatory signal corresponding to magnetization precession. Oscillation decay times vary with NM materials and Py thickness, indicating that $\alpha$ values are remarkably different between these films. The $\alpha$ values estimated using the Landau-Lifshitz-Gilbert (LLG) equation were 0.0087, 0.0145 and 0.055 for Ta/Py(20 nm)/Ta, Pt/Py(10 nm)/Pt and Pt/Py(2 nm)/Pt films, respectively [8]. TRMOKE data in the ps regime measured at $\theta_H=0^\circ$ for NM/Py/NM films is shown in Fig. 1(b). Here, signals were corrected by subtracting data measured at $H$=−4 kOe from that at $H$ = 4 kOe so as to exclude small non-magnetic contribution. TRMOKE signals for all films reach minimum values within 0.5 ps, which is longer than the laser pulse width. To estimate the demagnetization time quantitatively, the experimental data was fitted to the data calculated using the three-temperature model, which expresses the magnetization change as [1, 7]

$$
\Delta M(t) \propto [T_2 - T_1 \tau_E - T_2 \tau_M \exp(-t/\tau_M) + (T_1 - T_2)\tau_E \exp(-t/\tau_E)] \theta(t) \ast \Gamma(t). \tag{1}
$$

Here, $T_1$ represents the initial electron temperature rise, $T_2$ the equilibrium temperature, and $\tau_E$ the spin-lattice relaxation time. $\Gamma(t)$ is the Gaussian laser pulse profile, $\theta(t)$ is the Heaviside step function and $\ast$ signifies integral convolution of the these two functions. Heat diffusion and
Figure 1. (a) Time-resolved Magneto-optical Kerr effect signal for NM/Ni$_{80}$Fe$_{20}$ (Py)/NM films measured with applied magnetic field $H$ of 4 kOe at an angle $\theta_H$ of 23$^\circ$ between field direction and film normal, (b) Corrected TRMOKE signal for NM/Py/NM films with $H=4$ kOe at $\theta_H=0^\circ$. Solid curves correspond to calculated data which are fitted to experimental data.

The effects from state-filling are neglected in this study. Calculated data can be well-fitted to the experimental data, as shown in Fig. 1(b) with solid curves. The estimated $\tau_M$ values for NM/Py/NM films were $\sim$ 0.5 ps for all films. However, this result is inconsistent with the relation $\tau_M \propto \alpha^{-1}$ and the reason for this discrepancy is as yet unclear.

Fig. 2(a) shows an example of a TRMOKE signal for a CMS film annealed at 500$^\circ$C measured with applied magnetic field of 4 kOe at $\theta_H=20^\circ$. The magnetic field direction was in a plane between [001] and [110] for this CMS film. The magnetization precession is sustainable over 1 ns and the $\alpha$ value for this film was estimated to be around 0.002 using the LLG equation. This estimate is consistent with the earlier reported value measured by ferromagnetic resonance [12]. Fig. 2(b) shows the MOKE signal as a function of magnetic field measured at a delay time of $\sim$0.5 ps and $\theta_H=0^\circ$. The MOKE signal displays a hysteresis loop, a behavior which is different from the linear behavior observed in NM/Py/NM films (not shown here). Such loops can be considered to originate from quadratic-MOKE (QMOKE), that are inherent in some Heusler alloys depending on the degree of L$_2$$_1$ order [13]. Here, we assume that the MOKE signal for CMS films is a linear combination of polar-MOKE (PMOKE) and QMOKE, as shown in Fig.2 (c). The PMOKE contribution can be obtained in a similar manner employed for the TRMOKE signals by subtracting the measured signal at $H=-4$ kOe from that measured at $H=4$ kOe. Fig. 2(d) shows the corrected TRMOKE signals in the ps time regime for CMS films annealed at 500$^\circ$C. The signal shapes are very similar to those for NM/Py/NM films and are well-fitted by calculated data shown with solid curve. QMOKE signals from the CMS films increased with annealing temperature, which is consistent with increasing L$_2$$_1$ ordering with annealing temperature. However, the corrected TRMOKE signals in the ps time regime did not show significant differences and the estimated $\tau_M$ values were almost independent of annealing temperature. The average $\tau_M$ value evaluated by parameter fitting was $\sim$ 0.3 ps, which is slightly smaller than those observed in Py films and is nearly the same as the polycrystalline CMS value [2]. This result cannot be explained simply by a $\tau_M \propto (1-P)^{-1}$ relation because the values of $P$ are very different for our CMS and Py films. Further experiments to study temperature and laser wavelength variation might be necessary.
Figure 2. (a) Time-resolved Magneto-optical Kerr effect signal (TRMOKE) for Co$_2$MnSi (CMS) film annealed at 500°C measured with applied magnetic field $H$ of 4 kOe at $\theta_H=20^\circ$. $\theta_H$ is defined as the angle between field direction and film normal (CMS [001] orientation), (b) MOKE signal as a function of magnetic field measured at $\theta_H = 0^\circ$ and delay time of ~0.5 ps, (c) Schematic illustration of observed MOKE signal, Polar-MOKE and Quadratic-MOKE, (d) Corrected TRMOKE signal for CMS film annealed at 500°C with $H=4$ kOe at $\theta_H=0^\circ$. The solid curve is the calculated data obtained by parameter fitting to the experimental data.

4. Summary
We investigated ultrafast demagnetization and magnetic damping for NM/Py/NM (NM=Ta,Pt) and epitaxial CMS films using an all-optical pump-probe technique. The TRMOKE signals exhibited rapid decrease in the sub-ps time regime and damped oscillatory behavior. The $\tau_M$ and $\alpha$ values were evaluated using the three-temperature model and the LLG equation. The $\alpha$ values for NM/Py/NM films depended on Py thickness and NM materials, while $\tau_M$ was almost constant. The $\tau_M$ values for CMS films were independent of $L_2$-ordering and a little shorter than those for Py films, even though the spin polarization is close to unity.

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