# Enhancement of Excitation Efficiency of Photo-Excited Precession of Magnetization in Co/Pd Multilayers with Oblique-Angle Excitation

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#### Abstract

We first review our previous work on photo-excited precession of magnetization (PEPM) in Co/Pd multilayer under low-power excitation [1], and point out the enhanced efficiency of transferring excitation energy to a magnetic system with decreasing the thickness of Pd layer. We then show further enhancement of excitation efficiency with PEPM data obtained with oblique-angle excitation [2], and discuss that the observed enhancement is due to the suppression of ultrafast loss of excitation energy caused by the anisotropic ultrafast energy transfer between parallel and perpendicular directions with respect to the propagation direction of an excitation light beam.

## 1. Introduction

Optical excitation of a magnetic system with fs laser pulses gives us the opportunity of studying its ultrafast dynamic response and manipulation of magnetization [3]. PEPM has been proved to be a powerful tool to extract physical parameters which are critical to the development of spintoronics materials [4]. Such transient magnetization behavior has the functionality of GHz to THz polarization modulation via a magneto-optical (MO) effect [5], and furthermore, has potential for temporal memory functionality in optics/photonics [5, 6] which is free from power for memory retention. Efficient excitation of magnetization with low-power pulses is also required for device operation. With this in mind, we have been working on PEPM of Co/Pd multilayers with low-power excitation light pulses. In this presentation, we discuss the enhancement of precession amplitude in PEPM with two different approaches; (1) modulation of sample structure and (2) variation of excitation condition.

## 2. Experimental

Ultra-thin Co/Pd multilayers (MLs) are well known for their interface magnetism [7], by which magnetic anisotropy can be engineered between perpendicular and in-plane anisotropy by changing layer thicknesses of both Co and Pd layers. Samples were prepared at 150 °C by DC magnetron sputtering on the Pd (4.86 nm) / Ta (2.18 nm) binary seed layer deposited on a Si (110) substrate. Thickness in MLs were varied in the range  $t_{Co} = 0.32 \sim$ 1.03 nm for Co layers, and  $t_{Pd} = 0.81$  or 1.62 nm for Pd layers. See ref.1 for detail.

PEPM was studied at room temperature by

time-resolved magneto-optical (TRMO) Kerr rotation spectrometry using pump and probe technique. A mode-locked Ti: sapphire laser was used as the light source, whose photon energy, pulse duration, repetition rate, and fluency are 1.568 eV, 150 fs, 80 MHz and  $F = 11 \mu J/cm^2$ , respectively. An external magnetic field of 2000 Oe was applied on samples with the tilt angle 65° with respect to the axis normal.

### 3. Influence of changing the Pd layer thickness

Figure 1 (a) shows TRMO signals of five samples with different  $t_{Co}$  values with the constant value of  $t_{Pd} = 1.62$  nm. Clear precession signals were observed despite of relatively weak excitation. Fitting experimental data with a dumped sinusoidal equation,  $A = A_0 \exp(-t/\tau) \cdot \sin(2\pi ft + \phi)$  reveals that the precession amplitude  $A_0$  varies with  $t_{Co}$  whereas precession frequency *f*, lifetime  $\tau$ , and initial phase  $\phi$  hardly change. The observed variation in  $A_0$  are moderate for  $t_{Pd} = 1.62$  nm. However,  $A_0$  increases ten times when  $t_{Pd}$  is decreased in half,  $t_{Pd} = 0.81$  nm, as shown in Fig. 1 (b).



Fig. 1 (a) TRMO signals of five samples with  $t_{Pd} = 1.62$  nm, and (b) TRMO signals of two samples with  $t_{Pd} = 1.62$  and 0.81 nm ( $t_{Co} = 0.78$  nm).

TRMO signals in the ultra-short time domain (inset) exhibit spike-like features due to ultrafast demagnetization, from which we notice that the magnitude of demagnetization are comparable for these two samples. This fact suggests that the initial tip-off angle of magnetization, which determines the value of  $A_0$ , is not only governed by the magnitude of demagnetization. In other words, the data shown in Fig. 1(b) suggest the appearance of a new pathway which alters the magnitude of

interface-induced anisotropy in addition with conventional ultrafast demagnetization, when the Pd layer thickness becomes very thin.

## 4. Influence of excitation with oblique angle

Excitation of Co/Pd MLs with a light beam of oblique/grazing incidence can be regarded as a simple analogue to the excitation of a magnetic layer with light in a waveguide. For photonic excitation with oblique incidence, the shape of the excited area becomes anisotropic, which may influence the microscopic process of energy flow between electron and spin subsystems during the inducement of non-equilibrium magnetization as well as propagation of spin waves [8].

Shown in Figs. 2(a) and (b) are TRMO signals obtained from the sample with  $(t_{Co}, t_{Pd}) = (0.78 \text{ nm}, 0.81)$ nm) with various incidence angle of pump pulses. Probe pulses impinge nearly normal to the sample surface. To our surprise, PEPM can be induced with a large angle of incidence, being  $\theta_{pump} = 88^\circ$ , for *p*-polarization. Plots of  $A_0$ vs.  $\theta_{pump}$  are shown for both *s*- and *p*-polarized pump pulses in the inset of Fig. 2(c). One may suppose that the difference between s- and p-polarization may be understood qualitatively in view of polarization dependent reflection Rbased on Fresnel law; the absorbed energy in the MLs is written as  $E_{abs}(\theta_{pump}) = F (1-R)\cos\theta_{pump}$ . However, after correcting the difference in reflectivity, we find, through quantitative analysis with eq.(1), that the gradually increasing excitation efficiency  $\eta$  turns reduction at around  $\theta_{\text{pump}} = 55^{\circ}$  and returns one at  $\theta_{\text{pump}} = 65^{\circ}$  for the s-polarization, whereas it keeps increasing for the p-polarization, as shown in Fig. 2(c). See details of experimental conditions and analysis in ref. 2.

$$\eta(\theta_{pump}) = \frac{A_0(\theta_{pump})}{E_{abs}(\theta_{pump})} \left/ \frac{A_0(0)}{E_{abs}(0)} \right. \tag{1}$$

A scenario of the enhanced  $\eta$  value should be based on microscopic events occurring in the time domain of 1 ps, which we attribute to the lateral flow of excess energy driven by diffusion of photo-excited hot-electrons which causes spin flipping [9]. When the value of  $\theta_{pump}$  is small, the nearly circled pumped and probed areas overlap well to each other, and the energy flow to the outside of probed area is isotropic. When the value of  $\theta_{pump}$  becomes large, the area of excitation is elongated as shown on the bottom right of Fig. 2(c), which results in suppression of dynamic energy flow along the long axis of elliptic area due to the inflow into the probed area, and thus the enhanced modulation of interface magnetic anisotropy.

## 5. Conclusions

Efficiency of inducing PEPM in ultra-thin Co/Pd MLs have been discussed in terms of structure parameter of the sample and excitation geometry between light and the sample. Efficiency of excitation seems to be enhanced by reducing the thickness of Pd layer, and by applying oblique incident condition for pump pulses.



Fig.2 TRMO signals obtained with different incident angles of pump pulses (the inset) for *s*- (a) and *p*- (b) polarizations. (c) Plots of  $\eta$  vs. incident angle for *s*- and *p*-polarization. Inset shows plots of  $A_0$  vs.  $\theta_{pump}$  for two polarizations. Schematic illustration of the overlap between pump (white) and probe (gray) areas are also shown on the bottom part of the figure. Diameters are differed intentionally for the graphic clarity.

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