Ultrafast manipulation of spin and orbital angular momenta by light pulses

Takuya Satoh
(Department of Physics, Kyushu University)

All-optical magnetization switching has been studied extensively in recent years. A typical form of non-thermal magnetization control is the inverse Faraday effect (IFE). It involves rotation of the linear polarization of a probe pulse induced by a circularly polarized pump pulse in a transparent medium. Spin precession accompanied by the IFE has been reported by Kimel *et al*. in iron compounds [1)]. Spin precession is also observed with a linearly polarized pump pulse, in particular, a pulse polarized in a direction nonparallel to the crystal axes. This phenomenon is called the inverse Cotton-Mouton effect [2)].

The IFE has also been observed even in pure antiferromagnetic (AFM) NiO with no net magnetic moment in the ground state [3)]. The resonance frequencies of AFM materials reach the terahertz range, which is several orders of magnitude higher than that of FM materials. For that reason, AFM materials attract much attention in the context of ultrafast spin control. However, the mechanism of the observed spin oscillation by circularly polarized pulses remains unclear because of birefringence in the material. Here we discuss detailed mechanism using an NiO single domain which is optically isotropic [4)].

Moreover, we report on the observation of coherent spin oscillations in AFM CoO in a pump-probe experiment. The orbital momentum of the Co^{2+} ion is not fully quenched by the crystalline field. We show that spin-orbit interaction as well as exchange interaction plays an important role for low-lying magnetic excitation [5)].

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Ultrafast spin manipulation of sub-lattice magnetic system with light

Arata Tsukamoto
College of Science and Technology, Nihon University, Chiba 274-8501, Japan

The speed limits for magnetization reversal are of vital importance for spintronic devices, not only for storage media. For ultrafast manipulation of magnetization, optical laser pulses could serve as an alternative stimulus to trigger magnetization reversal. An ultrashort laser pulse allows excitation of magnetic systems at time scales much shorter than fundamental quantities such as spin precession or spin-lattice relaxation times. In particular, the laser excitation brings the magnetic medium into a strong non-equilibrium state[1)], where a conventional description of magnetic phenomena in terms of equilibrium thermodynamics and adiabatic approximations is no longer valid. Consequently ultrafast laser-induced magnetization dynamics is a new and rather unexplored topic at the frontier of modern magnetism.

Here our recent experimental studies of metallic multi-sublattice magnets are summarized. In particular, we focus on magnetization dynamics triggered by an ultrashort laser pulse in ferrimagnetic amorphous rare earth (RE)-transition metal (TM) alloys. The inequivalency of the magnetic sublattices, on the one hand, and a fine balance of their angular momenta on the other, lead to a very peculiar dynamic behavior. This becomes particularly obvious at short time scales, such as the appearance of a ferromagnetic-like state[2)] at time scales below a few picoseconds. The laser-induced ultrafast demagnetization of ferromagnets, already demonstrated in 1996[3)] to occur at a subpicosecond time scale, is still a subject of hot debate. Whether the angular momentum is dissipated into the lattice via phonons and defects, or whether it is carried away by hot electrons or the photons — are still questioned at the forefront of ultrafast magnetism. The element-specific XMCD measurements were performed[2)] to study transient regime of spin dynamics. In order to trigger ultrafast spin dynamics in GdFeCo alloy, the reversal of the magnetizations of the two sublattices is initiated by ultrafast heating of the sample using a 60 fs laser pulse in opposite orientations of the external magnetic field of 0.5 T. However, whereas the net magnetization of Fe has collapsed within 300 fs, the demagnetization of Gd takes as long as 1.5 ps. Remarkably, in spite of the strong antiferromagnetic (AFM) exchange coupling between the Gd and Fe sublattices, they apparently lose their net magnetizations independently, then surprisingly, within the time scale between the zero crossings of the Fe and Gd moments (that is, between 300 fs and 1.5 ps), the net Fe and Gd moments are aligned parallel along the z axis despite the AFM coupling of their spins in the ground state. This state is followed by an inter-sublattice relaxation of the angular momentum, leading to a deterministic switching of the magnetization driven by ultrafast laser-induced heating. We found deterministic magnetization reversal[4)] in same GdFeCo driven by an ultrafast heating of the medium resulting from the absorption of a sub-picosecond laser pulse without the presence of a magnetic field. From the theoretical discussion[5)], the reversal happens because of the interplay of these different demagnetization rates with the exchange interaction coupling the sublattices. These results demonstrate all-optical switching depends only on the amount of energy absorbed by the magnetic system, independent of the wavelength or helicity of the laser pulse. The role of the light helicity in this process is clarified as well. Because of different absorption coefficients for right- and left-handed circularly polarized light in GdFeCo, the switching threshold is helicity dependent. This explanation is consistent with all the experimental findings on all-optical light helicity-dependent magnetic switching[6)] so far, varying from single- to multiple-shot experiments.

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Laser induced THz spin dynamics in magnetic alloys

S. Mizukami[1], S. Iihama[2], Q. L. Ma[1], A. Sugihara[1], K. Suzuki[1], X. M. Zhang[1], Y. Ando[2], and T. Miyazaki[1]
([1]WPI-AIMR, Tohoku Univ., [2] Dept. of Appl. Phys., Tohoku Univ.)

Films of ordered magnetic alloys having uniaxial magnetic anisotropy show a perpendicular magnetic anisotropy, which are of quite importance for recent spintronics applications. On the other hand, a large perpendicular magnetic anisotropy causes very fast spin angular momentum prerecession because the Larmor frequency f is proportional to the magnetic anisotropy field H_k^{eff}. When magnetic films have both a large perpendicular magnetic anisotropy K_u^{eff} and small saturation magnetization M_s, those films can exhibit precession with frequency of f=100-1000 GHz owing to the large $H_k^{eff}=2K_u^{eff}/M_s$. This frequency region overlaps THz wave band, so that it could be expected that new phenomena are emerged from the mutual coupling between various THz excitations and spin dynamics. As a first step of exploring such new field, it is necessary to investigate materials exhibiting THz spin dynamics and its way of manipulation. THz spin precession can be accessed by ultrashort pulse laser, which is also interesting for terahertz wave applications from the practical points of view, such as ultrashort pulse laser driven GaAs THz emitter. While, there are few researches on THz spin dynamics in magnetic alloys, since there are not so much materials having enough large H_k^{eff}.

We, so far, reported various Mn based magnetic alloy films with a large perpendicular magnetic anisotropy of 5-15 Merg/cm^3, such as $D0_{22}$ Mn_3Ga [1], $L1_0$ MnGa [2], C38 MnAlGe [3], and $D0_{22}$ Mn_3Ge [4,5]. Those also have small magnetization of 100-500 emu/cm^3 as well as relatively small Gilbert damping constant. Fig. 1 shows the typical all-optical pump-probe time-resolved Kerr rotation measured in the MnGa epitaxial film using a pulse laser with low laser fluence. After strong ultrafast demagnetization at zero delay time, rapid precession is observed. Similar data were obtained in the other Mn-based alloys films and those frequency values of precession are summarized in Fig. 2 as a function of normal component of applied magnetic field. Precession frequency f linearly increases with increasing field, which was reasonably account as very fast Larmor precession owing to the large H_k^{eff}. The maximum f is 0.55 THz in case of Mn_3Ge epitaxial films. In order to obtain into the insight the physical mechanism behind the laser-induced precession, we investigated laser fluence and field dependence of dynamics and analyzed them using some physical models including thermally induced torque. The dynamics at low fluence region can be well explained by the calculation of one dimensional micromagnetic simulation based on the modified three temperature model taking into account of gradient of electron and lattice temperature (Fig. 1) [6].

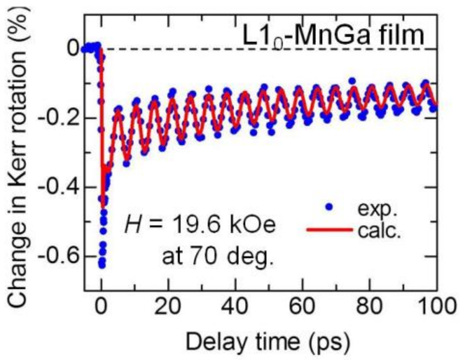

Fig. 1 Laser-induced spin dynamics of MnGa films. Solid curve is fitted to the experimental data.

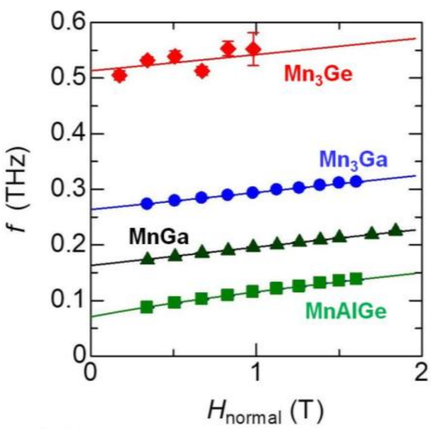

Fig. 2 Laser-induced spin precession frequency as a function of magnetic field for the various Mn-based alloy films. Solid lines are fitted to the data.

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Manipulation of ordered spins with light

– new photonic materials with magnetism –

H. Munekata
(Imaging Science and Engineering Laboratory, Tokyo Institute of Technology)

Owing to its high-speed, selective, non-linear, contactless, and quantum characteristics, light has the latent power of producing new functionality and paradigm, when combined with novel materials. This presentation reviews the frontier of research concerning a study of interaction between light and ordered spin (magnetization) with ultra-short light pulses and magnetic materials. A personal perspective as to new applications in the field of information processing and transmission is also discussed in view of photonics materials.

At the present stage, it is very important to establish reliable techniques for manipulating spins in magnetic materials with photons, and demonstrate prototype devices for mutual conversion between photons and ordered-spins. To this end, author, with his colleagues, studies experimentally the photo-excited precession of magnetization (PEPM) with various III-V ferromagnetic semiconductors [1,2] and metals [3] (Fig. 1), a concept of new photonic device consisting of those materials and existing optical components [4], and circularly polarized light emitters/detectors [5] (Fig.2). At the time of presentation, I plan to review experimental results on PEPM with Co/Pd ultra-thin multi-layers in the regime of weak excitation (< 1 μJ/cm^2) and a concept of all-optical signal modulation, added with experimental demonstration of electrical helicity switching.

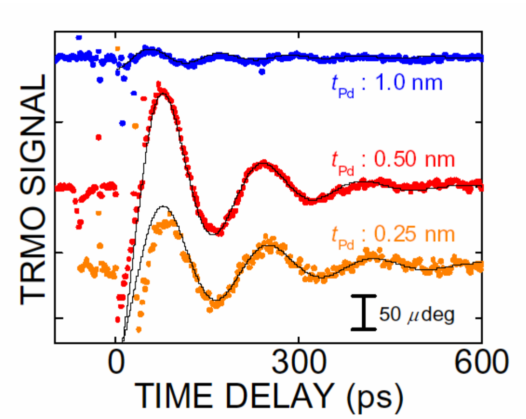

Fig. 1: Temporal profiles of PEPM data for three samples with different Pd layer thicknesses. Pump fluence 11 μJ / cm^2. The Co layer thickness was fixed at t_{Co} = 0.78 nm. Solid lines are fit to the experimental data.

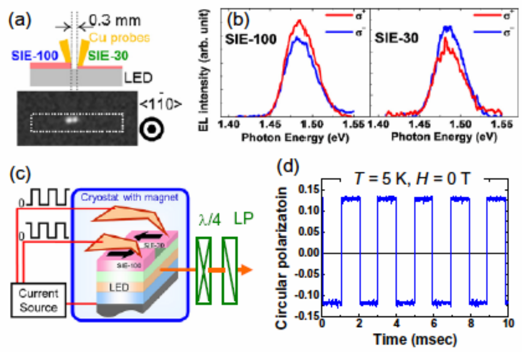

Fig.2: (a) a diagram of the spin LED with dual spin-injection electrodes (SIE) and its far-field image of EL emission, (b) EL spectra with current sent through each SIE, (c) a diagram of experimental setup, and (d) demonstration of electrical helicity switching at 1 kHz.

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Gilbert damping in magnetic multilayers with perpendicular anisotropy

T. Kato[1], K. Adachi[2], Y. Kusanagi[3], S. Okamoto[3], N. Kikuchi[3], O. Kitakami[3], S. Iwata[4]
[1] Department of Electrical Engineering and Computer Science, Nagoya Univ., Nagoya 464-8603, Japan
[2] Department of Quantum Engineering, Nagoya Univ., Nagoya 464-8603, Japan
[3] Institute of Multidisciplinary Research for Advanced Materials, Tohoku Univ., Sendai 980-8577, Japan
[4] Eco-Topia Science Institute, Nagoya Univ., Nagoya 464-8603, Japan

The magnetic materials with large perpendicular magnetic anisotropy (PMA) and low Gilbert damping constant are quite attractive since they not only have sufficient thermal stability but enable efficient writing in microwave assisted magnetic recording (MAMR) and spin transfer torque based magnetic random access memory (STT-RAM). Recently, we have studied the relationship between Gilbert damping α and PMA of Co-based multilayers, and reported that the α is closely related with thickness ratio of the multilayers, while almost independent of their PMA[1, 2], suggesting the possibility to obtain the multilayers with high PMA and low damping. In this talk, we summarize the Gilbert damping and anisotropy field of the Co-based multilayers which were evaluated independently by time resolved magneto-optical Kerr effect (TRMOKE) and coplanar waveguide ferromagnetic resonance (CPW-FMR), and discuss systematically the variation of the PMA and Gilbert damping with their layered structures.

Co / Ni, Pd, Pt multilayers with various layered structure were prepared on thermally oxidized Si substrates by a DC magnetron sputtering system. TRMOKE spectra were measured by pump-probe method using high-power fiber laser with λ = 1560 nm. During the measurements, an external field H_{ext} up to 8 kOe was applied in the direction of 45 deg from the film normal. For CPW-FMR measurements, rf current was fed into CPW by a vector network analyzer, and the complex scattering parameter S_{21} was recorded varying an rf frequency under an static field along film normal direction to estimate the resonance frequency f_{res} and linewidth Δf.

Figure 1 (a) shows t_{NM} / t_{Co} dependence of H_{keff} estimated from the TRMOKE and CPW-FMR measurements, where t_{Co} is the thickness of Co and t_{NM} is the thickness of Pt or Pd. Closed and open circles represent the data of the Co/Pt multilayers estimated by TRMOKE and CPW-FMR, respectively, and closed squares are the data of Co/Pd by TRMOKE. From Fig. 1 (a), the H_{keff} was confirmed to be roughly proportional to $1/t_{Co}$ at a constant t_{NM}. Although there are slight deviations between H_{keff} estimated from TRMOKE and CPW-FMR, overall tendency was similar to each other. The TRMOKE and CPW-FMR also show the similar results on t_{NM} / t_{Co} dependence of α as shown in Fig. 1 (b). The Gilbert damping α of the multilayers are proportional to t_{NM} / t_{Co} although the H_{keff} is independent of t_{NM} / t_{Co}. The linear tendency of α on t_{NM} / t_{Co} is considered to be explained by the spin pumping model[3]. The slope of α was dependent on the noble metal as shown in Fig. 1 (b). The Co/Pt has larger slope than Co/Pd, which may reflect the difference of the spin diffusion length between Pt and Pd.

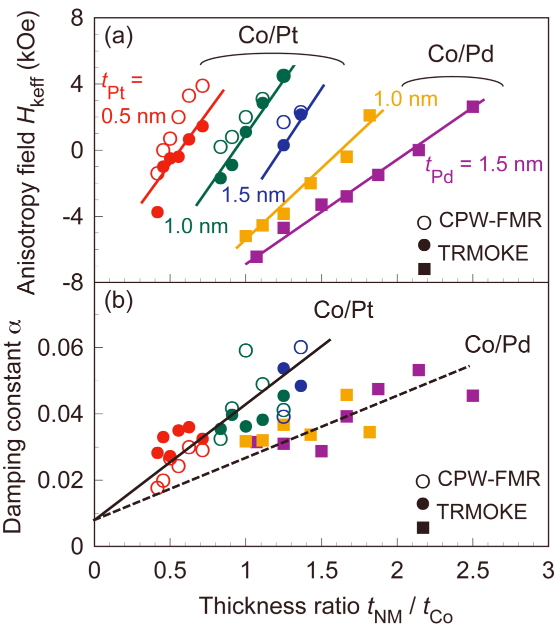

Fig. 1 Dependences of (a) H_{keff} and (b) α on the thickness ratio t_{NM}/t_{Co}. Closed and open symbols are the data estimated by TRMOKE and CPW-FMR, respectively.

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Surface Plasmon Techniques for Ultra-High Density Magnetic Recording

Katsuji Nakagawa
College of Science and Technology, Nihon University, Chiba 274-8501, Japan

Magneto-Optical (MO) recording is one of the primitive techniques to apply spin manipulation using light to change the temperature of magnetic films. MO recording itself is very simple to locally heat the magnetic film to change the magnetization *M*, but the mechanism of MO recording is more sophisticated than expected. The distribution of *M* caused by light absorption affects spin manipulation, such as Direct Over Write (DOW)[1], Magnetic Super Resolution (MSR)[2], MAMMOS[3], and 3D-MAMMOS[4], because each magnetic layer which has different characteristics on temperature is layered over a substrate. Some functional layers take roles by changing its stray field and/or exchange coupling between layers depending on temperature distribution in DOW, MSR, and MAMMOS.

Recently, we also use a heat technique on magnetic recording, so called Thermally Assisted Magnetic Recording (TAMR)[5]. Since the TAMR technique is just assisting magnetic recording by heat, it is simpler than the MO recording technique. The most improved key point in TAMR, compared with MO recording, is an extremely small spot size of light. Almost 10 nm in diameter is the spot diameter of light is the tentative goal for TAMR, even though sub-micron in diameter was the smallest spot size for MO recording. Sub-micron limit is caused by the diffraction limit for visible light, but we need around 10 nm spot in diameter for TAMR. Beyond the diffraction limit, we have chosen the near-field optical light which is generated by localized surface plasmon[6-9]. A light spot of 10 nm in diameter is available by applying the near-field. It was confirmed not only by simulation but also by the experimental result[10] that some magnetic domains were written by the local heat which was generated by surface plasmon antennas as well as femto-second laser.

How to deliver light power into a small plasmon antenna tip close to magnetic core is also an important key issue to create a heat spot on magnetic layer for TAMR. Some hybrid magnetic head systems with optics have been propoese[5, 11]. Planar Solid Immersion Mirror (PSIM)[11, 12] as well as plasmonic waveguide applying surface plasmon polaritons[11] have a high potential to effectively deliver light power in a hybrid magnetic head.

As the possibility that magnetization can be controlled by the helicity of ultra-short laser pulse was reported[13], the idea of all optical magnetic recording has been carried out. The method applying the helicity of light has a potential to accelerate magnetic recording speed, but its recording density is limited by the spot size of the circularly polarized light. Surface plasmon antenna for circularly polarized light was also studied[14]. It was revealed by a simulation that a circularly polarized light in a magnetic particle of 15 nm in diameter was able to be confined by a surface plasmon technique.

Spin manipulation using light is a new idea as well as an old idea. If we learned from the historical MO recording techniques, and applied them to current issues, we could improve the techniques beyond TAMR.

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Polarization Properties of a-SNOM

○ T. Ishibashi[1], Q. Meng[1], Y. Cai[1], S. Ikeda[1], H. Ono[1], A. Emoto[2], T. Shioda[3]

([1]Nagaoka Univ. Tech., [2]Doshisha Univ., [3]Saitama Univ.)

The aperture less scanning near-field optical microscopy (a-SNOM) is a promising technique to control spins using light in nano-scale, because a near-field light utilized in a-SNOM is enhanced several orders stronger than an incident light and could have a diameter of < 10 nm. However, controlling of polarization of near-field light in a-SNOM is the most important issue to be solved. In order to control the polarization of the light in the a-SNOM, we need to understand polarization properties of a-SNOM first.

The a-SNOM developed in this study is based on a commercial scanning probe microscope (SPI3800N probe station and SPA300 unit, Seiko Instrument Inc.)[1)]. A cantilever (SI-DF3P2, SII NanoTechnology Inc.) made of silicon having an extremity's radius of 7 nm with a resonant frequency Ω of ~ 80 kHz is used. A laser diode (TC20-4030-4.5/15, Neoark corp.) with a wavelength of 408 nm is used as a light source. The laser beam is focused on the top of the probe by using a plate-type lens with an incident angle of 45 degrees. A scattered light from the sample's surface in the near-field close to the tip apex is measured by a photomultiplier tube placed after the beam splitter. Signals were measured by the lock-in detection method for reference frequencies of Ω and 2Ω. Polarization properties were measured by using a set of polarizers. FDTD simulation was carried out to analyze distribution and polarization of lights around probes.

Figure 1(a) shows intensities measured for a Cr film plotted as a function of the angle of the analyzer θ_a, where the azimuth angle of the incident light, θ_i, is 40 degrees. We found that the shape is like a four-leaves clover, while a linearly polarized light gives a curve expressed by $\cos^2\theta$. We have decomposed the result by assuming that it is a superposition of signals from the top of the probe and the background as shown in Fig.1(b), where the background signals were calculated as an intensity of light reflected by a probe and a Cr film. Consequently, we obtained azimuth angles of SNOM signals for each θ_i as shown in Fig.2. A result of FDTD simulation is also plotted in Fig.2, which agrees well with the measured data. We also find that the a-SNOM act as wave plate expressed by Jones matrix, indicating that the polarization property of the a-SNOM is maintained.

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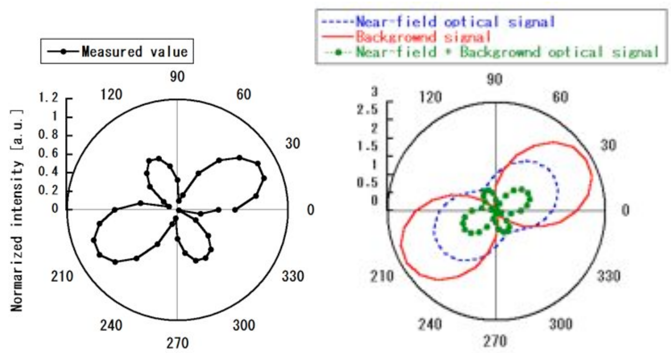

Fig.1 (a) Intensities measured for a Cr film with θ_i = 40º, plotted as a function of θ_a, (b) a result of fitting.

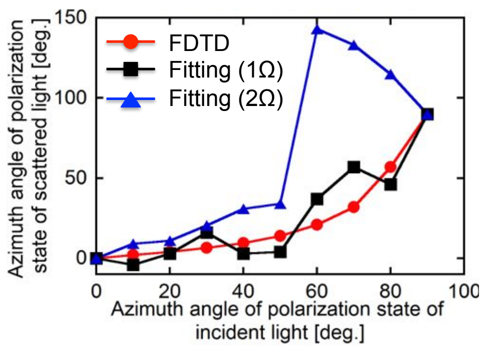

Fig.2 Azimuth angles of SNOM signals for each θ_i.

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