Ultrafast optical control of magnetization dynamics in ferrimagnet with antiferromagnet-like spin order

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Ultrafast manipulation and detection of spin dependent phenomena are crucial for future applications on magnetic and spintronic devices. Controlling magnetism by light is one of the promising approaches. For optical manipulation of magnetic systems, femtosecond laser pulses that are among the shortest stimuli with energy and angular momentum in contemporary technologies could serve as an alternative stimulus to trigger magnetization reversal and spin current etc. An ultrashort laser pulse allows excitation of magnetic systems at time scales much shorter than fundamental quantities such as spin precession or typical spin-lattice relaxation times. As an exciting and encouraging example, we reported that a direct demonstration of deterministic All-Optical magnetic Switching (AOS) was observed[1-3] in ferrimagnetic GdFeCo alloys in the absence of an external magnetic field at room temperature, which became subject of intense discussion in modern magnetism. In addition, in the similar material system, we have also reported an acceleration of magnetization dynamics originated from the angular momentum compensation phenomenon[4-5].

In this talk, I will introduce that excitation and time-resolved observation using ultrashort pulses of various wavelengths play an important role in elucidating the mechanisms of AOS and novel spin dynamics, and in pursuing their potential as magnetization control technology.

In pursuit of the mechanism of dynamic behavior in ferimagnet, it was desired to clarify the dynamics of each sublattice magnetization in a ferrimagnet having a sublattice magnetization structure with different main magnetic origins and having an antiparallel coupled spin order like an antiferromagnet. Time-resolved element specific measurements (X-ray magnetic circular dichroism) of the sublattice magnetization dynamics when AOS is excited by short pulse laser (visible light) reveal that the time-response characteristics of the magnetization differ between Gd and Fe, resulting in the emergence of a transient ferromagnetic state[6]. It was later found that having such a difference in antiparallel coupled magnetization response plays an important role in the mechanism of the deterministic AOS phenomenon. In addition, it was found that AOS can be excited even by pulsed THz light with low photon energy, and that the pulse length is one of the important driving conditions[7-8].

From a technological point of view, it is desired to minimize the light controllable spatial size.
After exciting AOS in a small area of TbFeCo by a gold two-wire antennas with short pulse laser (visible light), the magnetic behavior under the antenna structure was observed with time resolved resonant X-ray holography and magnetic circular dichroism[9]. It was shown that ultra-high-speed magnetization reversal can be achieved in a down scaling to about 50 nm by the AOS phenomenon. Furthermore, by actively utilizing the properties of light, layer-selective magnetization reversal control of multilayer magnetic films is also possible by controlling incident light polarization[10]. In this talk, the possibility of magnetic control and magnetic measurement using light will be introduced.

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Magnetization switching in Pt/Co/Pt multilayers by circularly polarized ultrashort optical pulses

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For thin magnetic metals, the magnetization is switchable by circularly polarized ultrashort laser pulses, referred to as all-optical helicity-dependent switching (AO-HDS). AO-HDS was first demonstrated in ferrimagnetic GdFeCo [1] and later in ferromagnetic metals, including multilayered Co/Pt and Co/Ni stacks and granular FePt medium [2]. Because state-of-art magnetic hard-disc drives use local heating by laser illuminations [3], the additional use of the optical helicity should be beneficial for further decreasing the energy consumption of writing a magnetic bit. However, the path toward the application is steep for difficulties, such as the slow dynamics, the necessity of multiple optical pulses, etc. Furthermore, it remains an outstanding issue that the dominant mechanism is unclear, i.e., which thermal or nonthermal effects dominate the AO-HDS.

In this symposium, I would like to present our recent progress in studies of the AO-HDS in ferromagnetic Pt/Co/Pt multilayers that are most typical for AO-HDS. The main driving force of the AO-HDS by multiple fs pulses was explored in the near-infrared to the visible spectral range. The helicity-dependent laser absorption by the magnetic circular dichroism predominantly accounts for the monotonic increase of the switching efficiency with increasing the wavelength from 0.5 μm to 1.2 μm [4]. We also found that a single pair of a fs linearly polarized pulse and a ps circularly polarized pulse with a time separation of a few picoseconds enables us to deterministically switch the spins in a helicity-dependent way on a picosecond time scale [5]. In the middle of these experiments, we were inspired to switch the magnetization by “x-ray” magnetic circular dichroism resulting from the core-to-valence electric-dipole transitions. Using the cutting-edge Japanese x-ray free electron laser SACLA, we discovered fs circularly polarized x-ray pulses control the magnetization when the photon energy is set at the absorption edge of Pt.

These projects were conducted in collaborations with Th. Rasing group (Radboud University), A. V. Kimel group (Radboud University), A. Kirilyuk group (Radboud University), T. Ono group (Kyoto University). M. Suzuki (Kwansei Gakuin University), Y. Tanaka group (University of Hyogo), I. Matsuda group (University of Tokyo), R. W. Chantrell group (University of York), and Advanced Light Source and Optics Research Group, XFEL Utilization Division (SACLA).

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Spin dynamics in ferromagnetic and antiferromagnetic thin films studied by ultrafast lasers

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Laser-induced reaction of magnetic materials is important in the development of magnetic data storage devices. A time-resolved microscope is a powerful experimental tool to study the dynamics of magnetic domains. We studied laser irradiated magnetic domains in $NiCo_2O_4$ thin films by using time-resolved magneto-optical Kerr effect (TR-MOKE) microscope [1]. Figure 1 shows the experimental setup of TR-MOKE microscopy with the pump-probe method. Femtosecond Yb:KGW laser Pharos (1030 nm, 1 kHz, FWHM ~ 0.2 ps) was used as a light source. The $NiCo_2O_4$ thin films were excited by horizontally polarized pump pulses (1030 nm) and observed by probe pulses, which had half wavelength (515 nm) based on the second harmonic generation. By using this microscope, we revealed that the value of the demagnetization time constant is about 0.4 ps, indicating ultrafast demagnetization. This time constant was significantly smaller than the large time constants reported for other half-metallic oxides, and agrees with the spin polarization of ~ 0.7 determined by tunnel magnetoresistance.

Ultrafast magnetization switching by pulsed lasers has been actively studied due to its potential for next-generation magnetic recording devices. A phenomenon where magnetization is switched by laser irradiation without applying a magnetic field is called all-optical switching (AOS). We are searching for AOS in oxide thin films to realize future device applications. By performing magneto-optical Kerr effect (MOKE) microscopy measurements of laser-irradiated magnetic domains, we revealed the accumulative-type AOS in $NiCo_2O_4$ thin films with a property of perpendicular magnetic anisotropy [2]. Here the sample was irradiated with linearly horizontal ultrafast laser pulses generated by Yb: KGW laser Pharos (1030 nm, 1 kHz, pulse width ~200 fs).

Figure 1 presents the schematics of the laser-pulse-accumulation effect of laser-induced magnetization switching in the $NiCo_2O_4$ thin film. AOS emerged at the perimeter of the lase spot after irradiating 10^3 - 10^5 pulses. Furthermore, the AOS area increased by accumulating laser pulses.

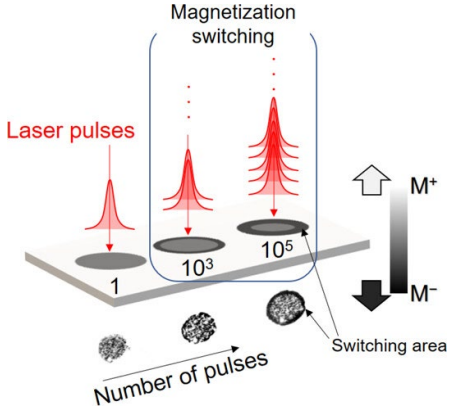

Fig. 1: Schematics of the laser-induced magnetization switching in the $NiCo_2O_4$ thin film [2].

Recently, we succeeded in building a low-cost and portable MOKE microscope device by using a 3D printer. This costs approximately 20,000 yen, much cheaper than standard commercial ones.

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Nonlinear and linear X-ray magnetic spectroscopy by ultrashort pulse X-ray lasers

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X-ray lasers, such X-ray free electron laser (XFEL) or high harmonic generations, have been developed and they generate ultrashort pulse, *i.e.* femtoseconds, with the wavelength of 100 nm ~ sub-nm [1]. The short pulse duration has allowed us to trace ultrafast spin dynamics by the magneto-optical experiment. Since the photon energy (hv) range covers the inner-shell absorption edges of materials, the signal carries the element or chemical information of a sample [2]. Thus, when we made a time-resolved measurement of such a resonance magneto-optical Kerr effect on the Co/Pt multilayer, we were able to layer-dependently trace magnetization dynamics of Co and Pt at hv = 60 eV (Co M-edge) and hv = 72 eV (Pt N-edge), respectively. The element selectivity can also be applied to the time-resolved experiment on the magnetic dopants in a sample [3]. We discovered transient enhancement of the magnetization of the Fe dopant in the semiconductor heterostructure induced by the optical pump [4].

Featuring XFEL, it is of note that this laser provides not only ultra-short pulses but also the ultrahigh intensity (peak brilliance). With the light source, materials science research using nonlinear spectroscopy such as Second Harmonic Generation (SHG) has become possible in the X-ray region [5-10]. An experiment of SHG itself has been studied for many years with infrared-visible lasers in the laboratory. X-ray experiments can again take advantage of optical transitions from atomic core levels to add elemental selectivity to the nonlinear signal. Therefore, it is possible to pinpoint the element or chemical species that constitutes the place where the inversion symmetry. In other words, it can become the "interface/surface selective" X-ray probe not the "interface/surface sensitive", as done with synchrotron radiation spectroscopy. This methodological feature is very powerful and we have succeeded in selectively capturing the microscopic behavior of lithium ions in lithium compound crystals and at the electrode interface of lithium battery [7-9]. Recently, we have also succeeded in observing magnetization-induced SHG (MSHG) in magnetic multilayer materials, $[Au/Fe/MgO]_n$, in X-ray region [10]. We found that the SHG signal at the resonance energy of the Fe M-shell absorption edge changes sensitively according to the direction of the magnetic field [10]. In the future, this nonlinear X-ray spectroscopy is expected to be of great use in academic and industrial fields as a new research approach in material science.

In the presentation, I introduce linear / nonlinear X-ray magneto-optical spectroscopy with our recent achievements and discuss the feature prospects of these methods [11].

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High throughput magneto-optical imaging and unconventional spin-wave dynamics

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Spin waves, which are fluctuations of ferromagnetic order, manifest diverse dynamics due to their intense nonlinearity and coupling with other excitations in solids. For the observation of the spin-wave dynamics, magneto-optical imaging has been a promising tool because it can obtain the spatial distribution of magnetization orientation, which has been already widely used to see magnetic texture such as magnetic domains and skyrmions.

In this presentation, we report the direct observation of various spin wave dynamics by using our newly developed time-resolved magneto-optical imaging method. One characteristic property of spin waves is their intrinsic strong nonlinearity, which leads to multi-magnon scattering processes of magnons, the quanta of spin waves. However, these magnon scattering processes, which occur during the manifestation of spin wave dynamics, have yet to be directly observed and have remained theoretical tools. In our newly developed time-resolved magneto-optical imaging method, we have succeeded for the first time in directly observing spin wave dynamics induced non-linearly by microwaves by synchronizing a microwave source and a pulsed laser for observation.

The presentation reports results on successful real-space imaging focusing on spin wave dynamics resulting from the parametric process, a process where one photon generates two magnons, and the four-magnon scattering, where two magnons scatter into two other magnons. We successfully clarify the localization of spin waves due to nonlinearity and the separation of contributions from multiple scattering processes.

In addition, magnons are considered to exhibit novel functionalities in the presence of the coupling between other elementary excitations. In general, when two oscillators are coupled, once oscillation is excited on the oscillator, the amplitude is transferred to the other oscillator in time, which comes back again after the same time it took to be transferred, a phenomenon called coherent oscillation. In a magnetic material, various elementary excitations are responsible for physical properties of the material, including the excitation of lattice and magnetic order, phonons and magnons. Owing to spin-orbit and dipole- dipole interactions, phonons and magnons are coupled to each other, which could lead to the coherent oscillation between magnons and phonons if the coupling is strong enough. In this talk, the experimental direct observation of the coherent oscillation between magnons and phonons are presented [1,2]. In a Bi-doped magnetic garnet, $Lu_2Bi_1Fe_{3.4}Ga_{1.6}O_{12}$, we observed coherent temporal oscillation between magnons and phonons as a result of hybridization, where magnons and phonons are coherently interconverted to each other during propagation.

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Observation of exchange bias switching using time-resolved-magneto-optical Kerr microscopy

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When a ferromagnetic layer is adjacent to an antiferromagnetic layer in a heterostructure, the hysteresis curve shifts as if the magnet feels an unidirectional effective magnetic field and the effective field is so-called the exchange bias. As one of its uses, field-free spin-orbit torque switching has been realized. In the ferromagnet/heavy metal heterostructures, the magnetization can be switched via spin-orbit torques (SOTs) when the in-plane external magnetic field is additionally applied [1]. However, when the antiferromagnetic layer is attached to the ferromagnetic layer, the external magnetic field can be replaced by the exchange bias and then field-free switching is realizable, which is useful from an application viewpoint [2].

Although exchange bias has been employed simply as the replacement of the in-plane external magnetic field, it has also been reported that SOTs switch the ferromagnetic spins as well as the out-of-plane component of the exchange bias [3]. Since the exchange bias arises from the interfacial antiferromagnetic spins, it is important to understand the temporal trajectory of the exchange bias switching regarding the high-frequency response of antiferromagnets [4]. We here investigated SOT-induced switching of both the magnetization and the exchange bias in Pt/Co/IrMn heterostructures using time-resolved magneto-optical Kerr effect microscopy (TR-MOKE) and micromagnetic simulation [5].

We deposited Si sub./Ti(5)/Pt(5)/Co(1.2)/IrMn(8)/Ti(2) (in nm) multilayers using magnetron sputtering at room temperature. The film was annealed at 200℃ in a magnetic field of 0.9 T, so the film has an exchange bias of 35 mT and 55 mT in the out-of-plane and in-plane axes, respectively. The film was subsequently patterned to 2×2 μm^2 square devices and Au microstrips were attached to the device as shown in Fig. 1a. Using a TR-MOKE setup and nanosecond-pulse currents (Fig. 1b), we experimentally realized time-resolved switching. As displayed in Fig.2a and b, we observed that the magnetization is stabilized at multiple levels and the levels depend on the in-plane external magnetic field and the current density. Comparing the obtained results to hysteresis loop experiments, it is found that the multilevel switching is linked to the exchange bias switching. Although the switching of the exchange bias has been reported using quasi-static measurements [3], our results indicate that the exchange bias is switched by SOTs at a rate equal to or faster than the magnetization.

To further understand the temporal trajectory of the exchange bias switching, we utilized MuMax3 for performing micromagnetic simulation [6]. The multilevel switching in the simulation behaves similarly to the experimental results and switching of the interfacial antiferromagnetic spins was observed (Fig. 2c-e). It implies that the SOTs indeed switch the exchange bias, and the multilevel switching occurs because magnetic domains in the Co layer are pinned by the interfacial IrMn spins. We will discuss the contribution of the Joule heating and the difference between each graph in detail in the presentation.

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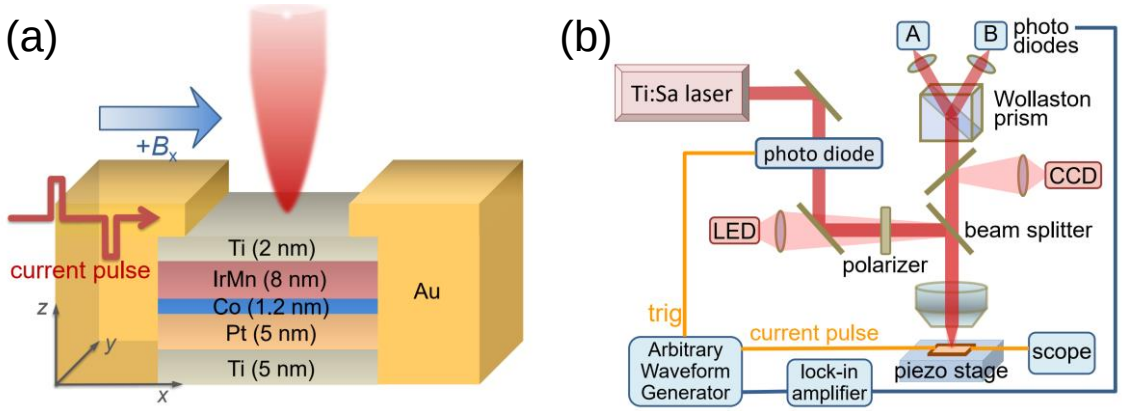

Fig. 1 (a) Schematic of the sample and measurement configuration. (b) Overview of the time-resolved magento-optical Kerr effect microscopy.

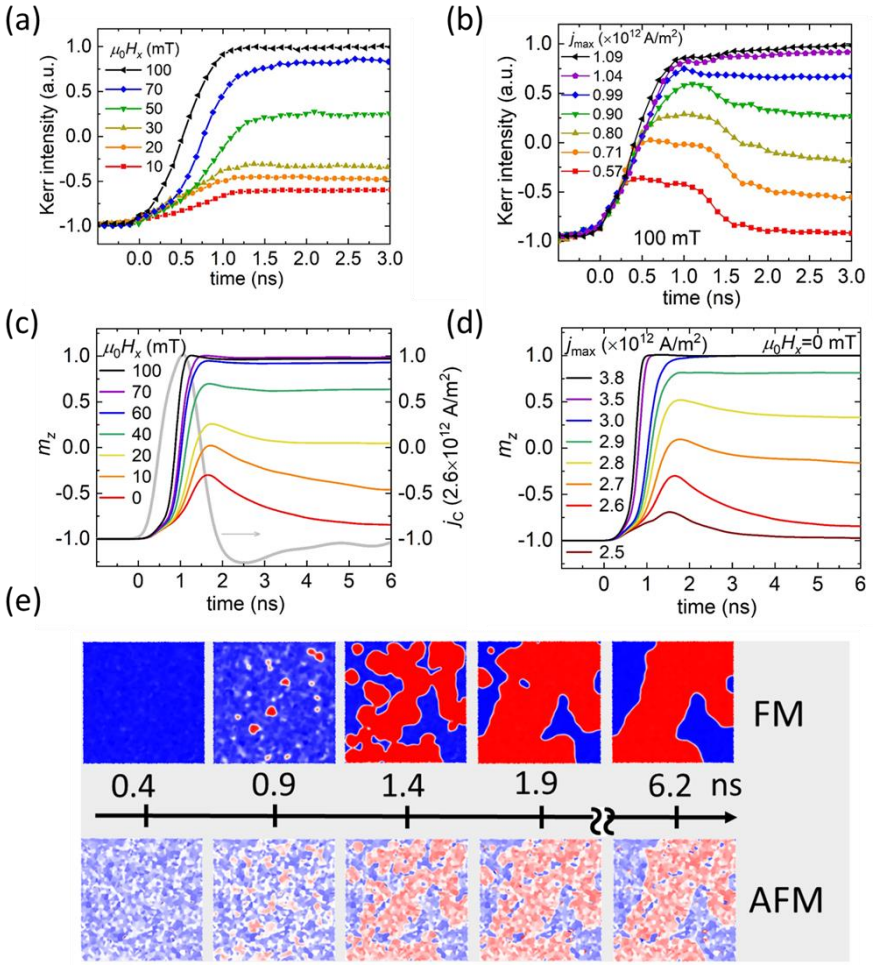

Fig. 2 (a,b) Experimental results of time-resolved SOT switching using (a) different magnetic field and (b) different pulse current density. (c,d) Simlation results of SOT switching as a function of time using (c) diffent magnetic field and (d) different pulse current density. (e) Time dependence of domain structures in Co layer (FM) and interfacial IrMn layer (AFM) obtained using micromagnetic simulation.