Ultrafast Dynamics and Control of the Spin Systems using Terahertz Magnetic Field

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Terahertz time domain spectroscopy (THz-TDS) based on femtosecond laser has become an established method for studying low-energy excitation dynamics. Especially, the usage of the magnetic field component of THz radiation was recognized as a powerful tool for investigating spin dynamics via magnetic dipole interaction. Up to now, we have demonstrated the applicability of this method to study spin states and dynamics in picosecond region for various materials [1-5]. This presentation aims to giving a brief overview of the study of spin manipulation with THz magnetic fields we have pursued so far, focusing mainly at the coherent spin manipulation in ferro-[1] and weak ferromagnets [2,3]. Specifically, here we show the capability of plasmonically-enhanced intense THz magnetic nearfield to coherent control of macroscopic magnetization in weak ferromagnet $ErFeO_3$ during spin reorientation phase transition (SRPT), which opens new route for the future spintronics devices.

The ever-growing demand for high speed spintronics devices propelled extensive searches of spin-manipulation methods based on ultrafast optical techniques, attempting to control the coherent spin dynamics and macroscopic magnetic order. Recent advancements in the THz time-domain spectroscopy techniques enabled direct visualization of ultrafast spin dynamics in the picosecond time scales. As the photon energy of THz region lies far below that of most electronic transitions, in this frequency region the magnetic field component of the THz pulse can directly access magnetic-dipole transition of electron spins without supplying excess heat into the system. In this presentation, we demonstrate some of our recent achievements of coherent control of macroscopic magnetization in spin systems using such THz magnetic fields. It is based on the plasmonic enhancement of magnetic fields in the nearfield region to locally amplify the spin precession motion, and the usage of an intrinsically magnetic fluctuation-sensitive process of SRPT.

The $ErFeO_3$ (001) single crystal, which is a canted antiferromagnet, was used as a sample. It shows SRPT between 85 and 96 K accompanying continuous orientation change of magnetization from *a*-axis to *c*-axis at low and high temperatures, respectively. The THz pulse with peak electric field up to 300 kV/cm were generated from $LiNbO_3$ crystal by the pulse front tilting method and used to pump a split-ring-resonator (SRR) structure fabricated on the sample surface. The spectral amplitude of the THz magnetic field was enhanced by over an order of magnitude by the SRR with a dimension of 200 x 200 μm^2 [4], to drive spin precession motion around the SRPT temperature (T=84K), which was probed by near-infrared (NIR) Faraday rotation. At the moment of spin precession we apply an NIR pulse to heat up the sample above SRPT temperature into the high-temperature phase. As a result of symmetry breaking of the directionality of SRPT caused by THz-induced spin precession, we succeeded in generating almost uniformly magnetized states in the final state, with its direction selectable by controlling the arrival timing of the heating pulse relative to the THz pulse (Fig.1). The final magnetization is over 80 % of saturation value. Our results indicate that even the small-amplitude spin precession can induce macroscopic change of spin order by utilizing phase transition process, extending the potential of THz application to controlling magnetic domains within picoseconds time- and subwavelength spatial resolution.

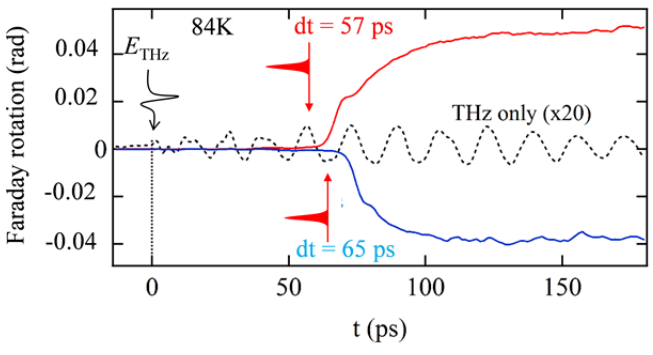

Fig. 1 Typical temporal evolution of out-of-plane magnetization measured by THz-pump optical Faraday probe experiment: Heating pulses arrive 57 ps (red) and 65 ps (blue) after THz irradiation at t = 0 ps. Black dotted curve shows spin precession without heating pulse.

References
[1] M. Nakajima, A. Namai, S. Ohkoshi, T. Suemoto, Optics Express **18**, 18260 (2010).
[2] K. Yamaguchi, M. Nakajima, and T. Suemoto, Phys. Rev. Lett. **105**, 237201 (2010).
[3] K. Yamaguchi, *et al*., Phys. Rev. Lett. **110**, 137204 (2013).
[4] T. Kurihara, *et al*., Phys. Rev. B **90**, 144408 (2014).
[5] K. Yamaguchi, *et al*., Phys. Rev. B **92**, 064404 (2015).

Optical-switching of second harmonic light in chiral photomagnet

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To control the physical properties and functionalities of materials via optical stimulation is an attractive issue. Spin-crossover phenomenon has been extensively studied because it realizes temperature-, pressure-, or photo-switching of the physical properties and functionalities. In particular, photo-switching from the low-spin (LS) state to the high-spin (HS) state, which is known as light-induced excited spin-state trapping (LIESST), is effective for optical control. Up to date, we have reported various unique photomagnetic mateirals using cyano-bridged bimetallic assemblies.[1,2] For example, we have reported an iron-octacyanoniobate metal complex, $Fe_2[Nb(CN)_8]\cdot(4\text{-pyridinealdoxime})_8\cdot 2H_2O$ and a photo-induced ferromagnetism originated by LIESST effect for the first time.[3] In this work, we synthesize a new 3-dimensional chiral cyano-bridged bimetallic assembly of iron-octacyanoniobate, $(\pm)\text{-}Fe_2[Nb(CN)_8](4\text{-bromopyridine})_8\cdot 2H_2O$ (**1**),[4] and firstly observed spin-crossover-induced second harmonic generation (SHG), light-reversible spin-crossover long-range magnetic ordering, and optical-reversible switching of the magnetization-induced second harmonic generation (MSHG) effect.

Cyano-bridged FeNb bimetallic assembly of **1** has a chiral structure in the $I4_122$ space group (Fig.1). The temperature (T) dependence of the molar magnetic susceptibility (χ_M) shows a thermal phase transition between the high-temperature (HT) phase and the low-temperature (LT) phase. The transition temperatures from the HT to LT ($T_{1/2\downarrow}$) and from the LT to HT ($T_{1/2\uparrow}$) are 112 K and 124 K, respectively. The UV-vis absorption spectra exhibits optical absorptions at 430 nm and 560 nm, which are assigned to $^1A_1 \rightarrow {}^1T_2$ and $^1A_1 \rightarrow {}^1T_1$ transitions on the Fe^{II}_{LS} site, respectively. Therefore, the transition from the HT to LT in the $\chi_M T$–T plot is due to spin-crossover from Fe^{II}_{HS} ($S = 2$) to Fe^{II}_{LS} ($S = 0$).

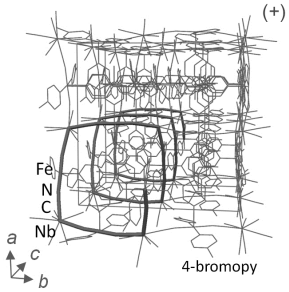

Fig. 1 Crystal structure of **1**.

Photomagnetic effect of **1** was investigated. Irradiating the LT phase with 473-nm light at 2 K produces large spontaneous magnetization. (Hereafter, called PI-1.) The magnetization (M) versus T curve shows a Curie temperature (T_C) of 15 K. The saturation magnetization (M_s) at 5 T is 7.6 μ_B, close to the expected M_s value of 7.8 μ_B due to ferrimagnetic coupling between Nb^{IV} (S = 1/2) and the photo-produced Fe^{II}_{HS} (S = 2). UV-vis spectrum and Mössbauer spectrum indicated that the observed bulk magnetization is due to the light-induced spin-crossover from Fe^{II}_{LS} to Fe^{II}_{HS}, i.e., LIESST effect. Next, we investigated the optical-switching effect on MSHG. Prior to irradiation, SHG for the LT phase of the paramagnetic state was measured. The SH intensity versus analyzer rotation angle (θ) plot shows that θ_{max} is 0° at $\pm H_0$, which is similar to the θ dependence of the SH intensity observed at 80 K. In the PI-1 phase, which is produced by LIESST effect with 473-nm light irradiation, θ_{max} at $+H_0$ is +88 ± 3° (Fig. 2). In contrast, at $-H_0$, θ_{max} is −86 ± 4°. In the PI-2 phase, produced by Reverse-LIESST effect with 785-nm light irradiation, the θ_{max} values are returned to +3 ± 1° and −3 ± 1° at $+H_0$ and $-H_0$, respectively. In the present system, LIESST and Reverse-LIESST effects control the polarization plane of the output SH light. The photo-reversibility was confirmed by alternative irradiation of 473-nm light and 785-nm light, which showed photo-reversible change in the SH intensity at θ = 0º.

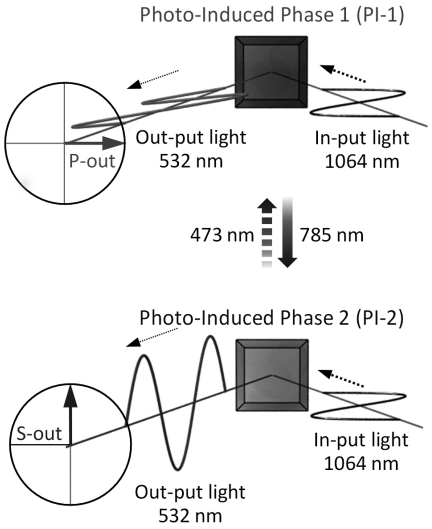

Fig. 2 Optical switching of the polarization plane of the output SH light between the PI-1 and PI-2 phases.

Reference
1) H. Tokoro and S. Ohkoshi, Dalton Trans., **40** (2011) 6825. 2) H. Tokoro and S. Ohkoshi, Bull. Chem. Soc. Jpn., **88** (2015) 227. 3) S. Ohkoshi, K. Imoto, Y. Tsunobuchi, S. Takano, H. Tokoro, Nature Chem., **3** (2011) 564. 4) S. Ohkoshi, S. Takano, K. Imoto, M. Yoshikiyo, A. Namai, H. Tokoro, Nature Photon., **8** (2014) 65.

Ultrafast optical excitation of magnetic materials

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The field of ultrafast opto-spintronics has emerged with a pioneering work on ferromagnetic Ni[1]. As one of the recent topics, we introduce our time-resolved spectroscopic studies on topological spin textures.

Magnetic skyrmions, spin vortices of topological origin, have been identified in several magnetic materials and at interfaces[2]. Their lateral extent depends on the magnetic interactions involved; some chiral magnets with the Dzyaloshinskii-Moriya interaction produce skyrmions with the size of 1-200 nm, which are suitable for magnetic information devices. These skyrmions show various emergent electromagnetic interactions, such as topological and Skyrmion Hall effects[2], and can be driven under an extremely small charge current or by magnons. The optical response/control of this nano-magnetic structure, in an ultrafast manner, will be of practical importance.

We report real-time dynamics of skyrmions in an insulating ferrimagnet Cu_2OSeO_3 [3], studied by all-optical spin-wave spectroscopy. The spins in the helical, conical, and skyrmion phases were excited by the impulsive magnetic field of the inverse Faraday effect[4] at non-absorbing photon energies, and subsequent precessional relaxations were detected through conventional magneto-optics. Clear dispersions of the helimagnon were observed, which was accompanied by a transition into the skyrmion phase when sweeping temperature and magnetic field. In the skyrmion phase, three collective excitations were identified, distinct from those in the surrounding conical phases (Fig. 1). These spin dynamics can be readily assigned to the clockwise/counter-clockwise rotations and breathing modes of the skyrmion crystal.

In addition to identifying the dynamics of topological spin textures, we are able to infer the spatial propagation and interaction of the magnetic excitations in these nano-magnetic spin structures by using variety of optical/microscopy techniques. As an example, we demonstrate an optical-drive of magnetic bubbles, which can be topologically equivalent to the skyrmion. In this case, magnetoelastic waves, coupled propagations of magnon and phonon, were photoexcited in iron garnet films. These magnetic excitations were found to interact with magnetic domains and domain walls more efficiently when the domain wall has steeper curvature[5] (Fig. 2).

If time allows, we will also discuss the spin-polarized photocurrent generation in ferromagnetic topological insulators, in which the mass-gap in the Dirac dispersion can be controlled by an external magnetic field.

Reference

1) E. Beaurepaire *et al.*, Phys. Rev. Lett. **76,** 4250 (1996).
2) N. Nagaosa and Y. Tokura, Nature Nanotechnol. **8,** 899 (2013).
3) N. Ogawa, S. Seki, and Y. Tokura, Sci. Rep. **5,** 9552 (2015).
4) A. Kirilyuk, A.V. Kimel, and T. Rasing, Rev. Mod. Phys. **82**, 2731 (2010).
5) N. Ogawa *et al.*, Proc. Natl, Acad. Sci. **112,** 8977 (2015).

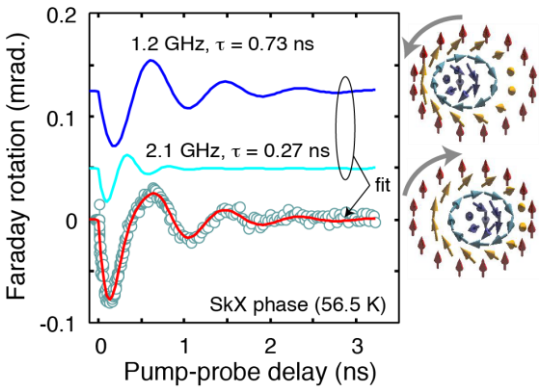

Fig.1 Spin precessions in skyrmion crystal phase.

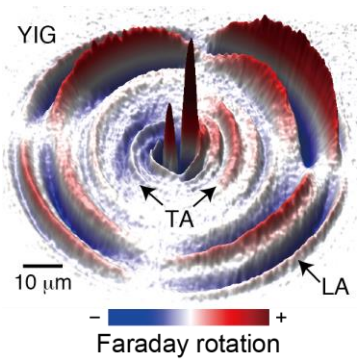

Fig.2 Optically-excited magnetoelastic wave.

Photoinduecd spin-state dynamics in thin film of perovskite cobalt oxide investigated by time-resolved x-ray diffraction

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Research on macroscopic quantum systems, which involve interplays among charge, spin, and orbital degrees of freedom, is important in condensed matter physics because it can lead to the emergence of unique optical, magnetic, elastic, and thermoelectric phenomena. The phase transition caused by optical stimulation is called photoinduced phase transition (PIPT) and has been of current interest during the past decades. With time-resolved techniques by using femto- and picosecond pulsed lights, it is possible not only to observe the dynamics of the electronic interactions among the degrees of freedom but to control the electronic, magnetic, and orbital order on an ultrafast timescale.

Perovskite-type cobalt oxides showing spin-crossover phenomenon have been regarded as promising materials exhibiting novel PIPT phenomena [1-3]. The $LaCoO_3$ containing the nominally trivalent Co ion (Co^{3+}) is a typical spin-crossover system and may take three different spin state: the low-spin (LS) state (t_{2g}^6, S = 0), intermediate-spin (IS) state ($t_{2g}^5e_g^1$, S = 1), and high-spin (HS) state ($t_{2g}^4e_g^2$, S = 2). The spin-crossover transition from nonmagnetic LS ground state to paramagnetic HS and/or IS states occurs with increasing temperature around 90 K. The recent research has revealed that the magnetic properties and the Co-3*d* orbital ordering can be tuned by the epitaxial strains [4,5]. In this study, we investigated the photoinduced state in an epitaxial thin film of $LaCoO_3$. To identify the transient structural change correlated with the spin states directly, we measured a time-resolved x-ray diffraction at PF-AR NW14A.

The single crystalline film (60 nm thick) of $LaCoO_3$ was fabricated on the $(LaAlO_3)_{0.3}(SrAl_{0.5}Ta_{0.5}O_3)_{0.7}$ (LSAT) substrate with (110) orientation by pulsed laser deposition, which exhibits the strain induced spontaneous magnetization (T_c = 94 K) and the lattice distortion with Co-3*d* orbital ordering below 124 K [4]. Figure 1(a) shows the (620) fundamental reflection profiles in $LaCoO_3$. The peak, which shows a single-peak feature at 120 K, splits into two at 80 K. The peak splitting is ascribed to the lattice distortion with Co-3*d* orbital ordering [4]. Figure 1(b) shows the difference peak profile between before and after photoexcitation at 80 K, obtained by an optical-pump and x-ray-probe technique. The x-ray pulse duration is 100 ps and the photon energy of the optical pump pulse is 3.10 eV with the second harmonics of the fundamental Ti:sapphire regenerative amplified pulse (photon energy: 1.55 eV, pulse duration: 120 fs, and repetition rate: 1 kHz). The shape of the difference peak profile indicates that the two peaks are changed toward the single-peak feature. This result suggests the structural change due to the photoinduced melting of the orbital order. In this talk, we will also present the transient reflectivity change reflecting the change of the spin state, measured by femtosecond time-resolved reflection spectroscopy, and discuss the PIPT in terms of the spin-structural dynamics.

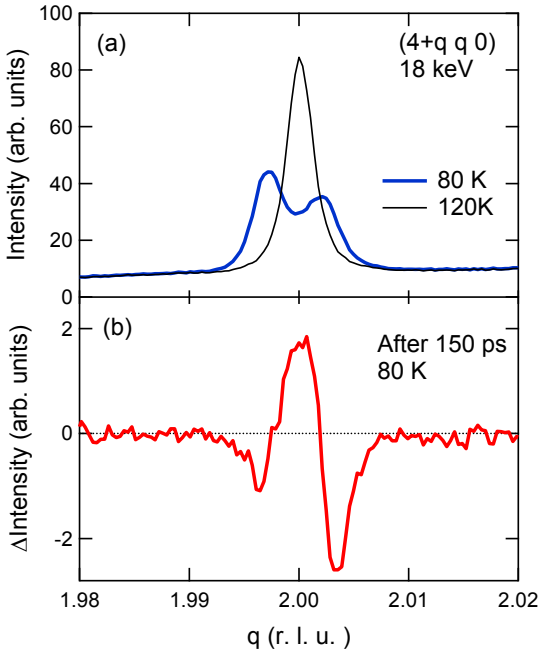

Fig. 1(a) X-ray diffraction profiles of (620) fundamental reflection along (110) direction at 80 K and 120 K. (b) Differential profile of fundamental reflection before and after photoexcitation (150 ps) at 80 K.

Reference

1) Y. Okimoto *et al*., Phys. Rev. Lett. 103, 027402 (2009).
2) Y. Okimoto *et al*., Phys. Rev. B 84 121102(R) (2011).
3) Y. Okimoto, R. Fukaya *et al*., J. Phys. Soc. Jpn. 82 074721 (2013).
4) J. Fujioka *et al*., Phys. Rev. Lett. **111**, 027206 (2013).
5) J. Fujioka *et al*., Phys. Rev. B **92**, 195115 (2015).

Magnetic dynamics study by soft X-ray photoemission electron microscopy

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By combining synchrotron radiation soft X-rays with Photoemission electron microscopy (PEEM), contrast maps based on X-ray absorption fine structures (XAFS) as well as X-ray magnetic circular/linear dichroism (XMCD/XMLD) can be obtained with the spatial resolution in the order of 10-100 nm.

In SPring-8, we possess two PEEM machines. While a spectroscopic photoemission/low-energy electron microscope (SPELEEMIII, ELMITEC GmbH) equipped in BL17SU beamline is widely used for electronic- or magnetic-states analysis of a variety of materials, taking advantage of high spatial resolution and various measurement functions (e.g. nano-XAS, micro-XPS and micro-LEED), a compact and versatile PEEM (PEEMSPECTOR, ELMITEC GmbH) in BL25SU plays an effective role in time-resolved imaging by pump-probe method (time resolution 50-100 ps). So far, we have performed dynamics analysis of magnetic vortex cores excited by pulses or radiofrequencies[1,2], ultrafast magnetization switching dynamics under the excitation by femtosecond laser pulses[3], and so on. Particularly in magnetic dynamics of ferrimagnetic GdFeCo films by pulsed laser excitation, we discovered giant propagating spin waves whose gyration angle reaching ±20°, given that the damping parameter of the magnetizations is suppressed and gyrotropic motion lasts long (that is to say, under the condition not appropriate for "ultrafast" magnetization switching)(Fig.1). These spin waves are suggested to be created as a consequence of strong spin-phonon coupling and further analysis is now in progress.

We are also developing new measurement systems for advanced spectroscopic and dynamics studies. For example, a prototype of complete co-axial sample cartridge for PEEM showed transmission capacity up to 5 GHz. This system is expected to be utilized for e.g. domain wall dynamics analysis or element-selective nano-FMR.

In this talk, principle of time-resolved PEEM using synchrotron radiation soft X-rays and examples of utilization researches as well as recent status of system development will be reviewed.

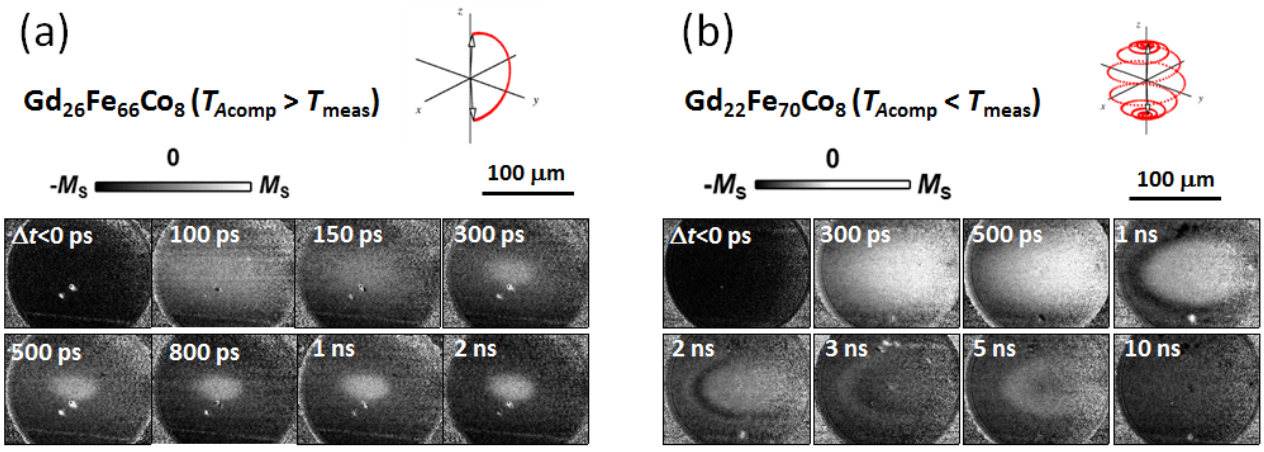

Fig.1 Time resolved Gd_M_5-edge XMCD-PEEM images of GdFeCo films promptly after pulsed laser excitation. (a) swift magnetization reversal (condition of strong precession damping) and (b) long-range spin wave propagation (prolonged spin gyration).

References

1) K. Arai et al. Jpn. J. Appl. Phys. 50 (2011) 053001.
2) T. Ohkochi et al. Jpn. J. Appl. Phys. 51 (2012) 128001.
3) T. Ohkochi et al. Jpn. J. Appl. Phys. 51 (2012) 073001.

Ultrafast dynamics studied by time-resolved x-ray diffraction

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Control of magnetic states by optical excitations in magnetically ordered materials has attracted considerable attention since the demonstration of ultrafast demagnetization in Ni within 1 ps, explored by time-resolved magneto optical Kerr effect studies 1). For this study, we chose time-resolved x-ray measurements to study ultrafast spin dynamics in transition-metal oxides.

X-rays from synchrotron radiation (SR) have time structures related to the SR pulse width of several 10 ps. X-ray free electron laser (XFEL) creates intense ultra-short (fs) x-ray pulses, enabling much more detailed study of the dynamics of the materials. We performed a time-resolved x-ray diffraction and scattering study in a pump-probe setup by using XFEL in LCLS (USA) and by using SR in BESSY (Germany). The pump light is Ti:sapphire laser (800 nm), and the probe is XFEL or SR.

Figure 1 (a) shows the time evolution of the intensity of the superlattice re ection (2 1/2 0) in charge and orbital ordered $Pr_{0.5}Ca_{0.5}MnO_3$ thin films 2). One can see clear oscillations, which correspond to the frequency of coherent phonons. Figure 1 (b) shows the time-resolved x-ray magnetic circular dichroism (XMCD) intensity in ferromagnetic $BaFeO_3$ thin films, showing rather slow demagnetization of ~ 100 ps 3), but this time scale was much faster in higher-fluence regions, suggesting photoinduced insulator-metal transitions.

This work has been performed in collaboration with P. Beaud, U. Staub, T. Tsuyama, N. Pontius, C. Schussler-Langeheine, M. Nakamura, S. Chakraverty, H. Y. Hwang, M. Kawasaki, and Y. Tokura, and has been supported by the Ministry of Education, Culture, Sports, Science and Technology of Japan (X-ray Free Electron Laser Priority Strategy Program).

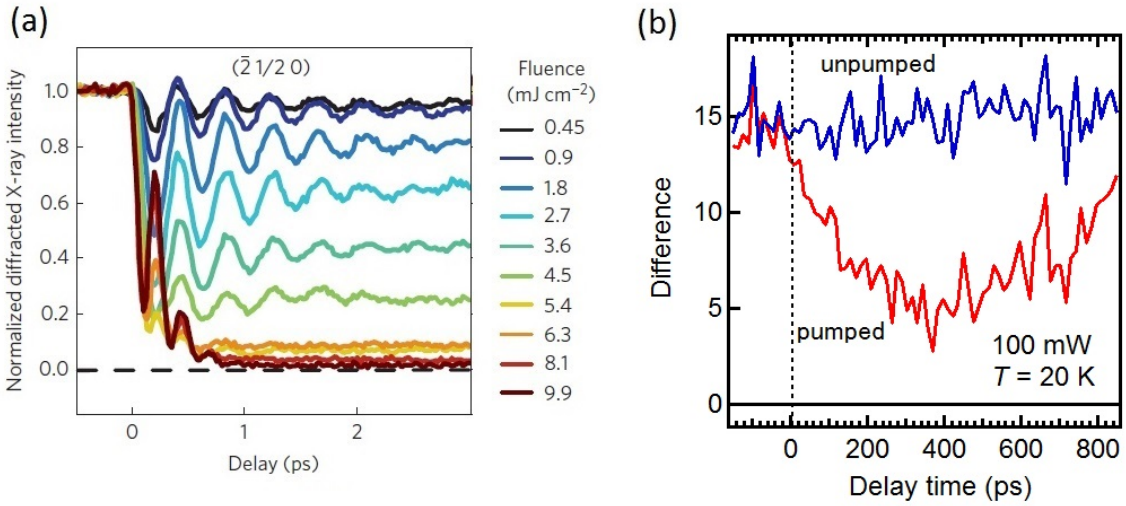

Figure 1: (a) Time evolution of the normalized diffracted x-ray intensity for the (2 1/2 0) reflection in $Pr_{0.5}Ca_{0.5}MnO_3$ thin films taken at 6.53 keV (off resonance). This superlattice peak is sensitive to the structural atomic motion. (b) Time evolution of the XMCD intensity in $BaFeO_3$ thin films taken at 710 eV (Fe $2p_{3/2}$ edge).

Reference

1) E. Beaurepaire *et al.*, Phys. Rev. Lett. **76**, 4250 (1996).
2) P. Beaud, H. Wadati *et al.*, Nat. Mater. **13**, 923 (2014).
3) T. Tsuyama, H. Wadati *et al.*, arXiv:1511.03365v1.