Ultrafast demagnetization of Pt magnetic moment in L1₀-FePt probed by magnetic circular dichroism at a hard x-ray free electron laser

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Abstract

Unraveling the origin of ultrafast demagnetization in multisublattice ferromagnetic materials requires femtosecond x-ray techniques to trace the magnetic moment dynamics on individual elements, but this could not yet be achieved in the hard x-ray regime. We demonstrate here the first ultrafast demagnetization dynamics in the ferromagnetic heavy 5d-transition metal Pt using circularly-polarized hard x-rays at an x-ray free electron laser (XFEL). The decay time of laser-induced demagnetization of L1₀-FePt is determined to be τPt = 0.61 ± 0.04 ps using time-resolved x-ray magnetic circular dichroism at the Pt L₃ edge, whereas magneto-optical Kerr measurements indicate the decay time for the total magnetization as τtotal < 0.1 ps. A transient magnetic state with a photo-modulated ratio of the 3d and 5d magnetic moments is demonstrated for pump-probe delays larger than 1 ps. We explain this distinct photo-modulated transient magnetic state by the induced-moment manipulation mechanisms as well as possible avenues for practical applications.

1. Introduction

Photo-induced magnetism has recently attracted significant attention because it reveals non-trivial spin manipulation mechanisms as well as possible avenues for practical applications [1–3]. An early time-resolved magneto-optical Kerr effect (trMOKE) study demonstrated an unexpected, ultrafast demagnetization of a ferromagnetic Ni foil within 1 ps [4]. Since then, extensive investigations have been conducted to find further novel photo-induced magnetic phenomena and explain their mechanisms [5]. One of the studied phenomena is the distinct, element-specific demagnetization behavior observed in multisublattice materials, as e.g. in ferrimagnetic GdFeCo [6]. Another topic of intensive research is all-optical helicity-dependent switching which
was first observed in ferrimagnetic GdFeCo alloy with a magnetization compensation point near room temperature [7]. Recent investigations demonstrated that helicity-dependent all-optical switching is possible even for ferromagnetic materials such as Co/Pt thin films and granular L1\textsubscript{0} FePt recording medium [2, 3].

To understand the mechanism of photo-induced magnetization dynamics and switching, element-specific measurements are necessary because magnetic materials that show remarkable photo-induced behaviors contain more than one magnetic elements [2, 3, 6]. X-ray magnetic circular dichroism (XMCD) in the soft x-ray regime is an element-specific magnetism measurement technique [8, 9] and time-resolved x-ray magnetic circular dichroism (trXMCD) can thus provide information about element-specific dynamics of magnetic materials. This technique has been applied to probe the element-dependent demagnetization dynamics at the 3d transition-metal L edges and rare-earth M edges in a number of multilattice materials and alloys [6, 10–13]. Alternatively, femtosecond pulses with energies in the extreme ultraviolet (EUV) range have been employed to trace the orbital- resolved magnetization dynamics of Gd [14] and the element-dependent magnetization dynamics of 3d alloys [15, 16] and trilayer systems [17]. In rare-earth-transition-metal ferrimagnets there is a compensation temperature which affects the photo-induced demagnetization processes. The role of the spin moment on the 5d orbitals in 4f metals were discussed recently [14, 18]; Frietsch et al report that the 4f and 5d spin dynamics differ in gadolinium metal [14]. In Gd there is a large 4f moment and a smaller 5d spin moment which is coupled to the 4f spin via intra-atomic exchange coupling. To study the complete spin dynamics the 5d spin dynamics needs to be probed as well. The EUV range has also been used to investigate recently the spin dynamics of composite ferromagnetic films as e.g. CoPt, NiPd, and FePt [19–21]. These studies indicated a small or almost no difference in the demagnetization behavior of the ferromagnetic element (Co, Fe, Ni) and the nonmagnetic element (Pd, Pt), but the signals at the Pt O edge or Pd N edge were not sufficiently large to determine a precise demagnetization mechanism.

To directly observe the dynamics of the Pt magnetic moment it is important to use an element-specific probe such as trXMCD that probes exclusively the spin-polarized 5d states. Thus far, the L\textsubscript{2,3} (2p→5d) edge of 5d transition metals, which lies in the hard x-ray region, could not be used for trXMCD, and therefore the element-specific spin dynamics of ferromagnetic materials with Pt have not yet been clarified. X-ray free electron lasers (XFELs) have recently enabled time-resolved measurements of ultrafast phenomena with ultrashort x-ray pulses with a duration of < 100 fs [22, 23] and have been applied to various research fields including magnetization dynamics [24, 25] and the dynamics of a 5d antiferromagnetic material observed by diffraction at an antiferromagnetic Bragg peak [26]. In this study, we report the first hard x-ray trXMCD measurements of the spin dynamics of the Pt moment in FePt using the XFEL with an x-ray phase retarder to produce circular polarized x-rays [27]. X-ray phase retarders have been widely used in several synchrotron radiation facilities such as the advanced photon source [28], European Synchrotron Radiation Facility [29, 30] and SPring-8 [31]. However, circularly polarized XFEL generation by an x-ray phase retarder is used only at SACLA; for trXMCD we developed a new measurement system utilizing an x-ray phase retarder and pumping laser. Doing so, we successfully determine the photo-induced ultrafast spin dynamics of Pt magnetic moments in L1\textsubscript{0} FePt thin films with perpendicular magnetic anisotropy; remarkably, we find the unfolding dynamics to be distinctly different from the ultrafast dynamics of the Fe moment. In addition to exhibiting all-optical switching, FePt has a high uniaxial magnetic anisotropy when it forms in the L1\textsubscript{0} structure and L1\textsubscript{0}-FePt is considered to be a candidate material for magnetic recording media, which is the reason why we chose the L1\textsubscript{0}-FePt as sample.

2. Results

Figure 1(a) shows the experimental setup for the pump–probe trXMCD based on the fluorescence yield method in SACLA BL3 [23]. Previously, time-resolved hard x-ray absorption spectroscopy [32, 33] and diffraction [34–36] have been reported from this beamline [37]; however, thus far, time-resolved x-ray absorption measurements with circular polarized XFEL pulses have not been performed. To combine the diamond x-ray phase retarder [27], the arrival timing-monitor system [38–40] and pump laser, we adopted the following optics setup: a pink (quasi-monochromatic) self-amplified spontaneous emission radiation with bandwidth of ~60 eV illuminated the diamond crystal tuned to generate circular polarization at a target x-ray energy, and the four-bounce channel-cut monochromator down stream [41] was used to select the monochromatic beam with circular polarization. We confirmed that the setup with a diamond crystal with thickness of 1.5 mm and Si(111) channel-cut crystals worked well to produce circular polarization with a high degree of circular polarization of \( P_\text{c} > 0.9 \) at 11.567 keV [42]. The arrival timing-monitor system [38–40] effectively compensated the jitter of XFEL and synchronized optical laser pulses and a resulting temporal resolution below 50 fs was achieved. The diamond phase retarder and the four-bounce channel cut monochromator decreased the XFEL intensity. We furthermore needed to set a smaller binning width for getting higher time resolution, which led to a decreased
signal per each delay point because the time-resolved results were obtained by binning the data according to the timing of the probe pulse. Nevertheless, our trXMCD setup worked well as shown below.

Figure 1(b) depicts the investigated element-selective spin dynamics. We used single-crystal L10-FePt thin films with a thickness of 20 nm on MgO(100) as samples. The Fe and Pt moment on the two sublattices are ferromagnetically coupled to one another in the L10 ordered structure. Photo-adsorption of an optical laser pulse creates ultrafast demagnetization on both Fe and Pt sublattices. To capture the transient dynamics of the Pt moment we measured the trXMCD at the Pt L edge with XFEL hard x-ray radiation.

The x-ray fluorescence detection method was used to acquire x-ray absorption and XMCD signals using a multiport charge-coupled device (MPCCD) detector [43]. The circularly polarized x-ray beam at the Pt L3 edge (≈11.6 keV) was incident on the sample in the direction normal to the film plane at a repetition rate of 30 Hz. X-ray fluorescence emitted from the sample was photon-counted and energy-analyzed by the MPCCD placed at the side with a distance of 150 mm. A Zn filter with a thickness of 50 μm was used to eliminate the strong elastic and Compton scattering from the sample substrate. According to x-ray fluorescence spectra, the Pt Lα fluorescence was observed around 9.4 keV as shown in figure 1(c). X-ray absorption signals of Pt were measured as the photon counts integrated over an energy window of 9.15–9.83 keV. The magnetization of the sample was saturated with a perpendicular magnetic field of 0.6 T applied using a permanent magnet (see appendix). A Ti:sapphire laser with a photon energy of 1.5 eV with pulse duration of approximately 30 fs was used to pump.

We show the results of x-ray absorption and the XMCD spectrum of L10-FePt using our setup in SACLA. Figure 2(a) shows the x-ray absorption at the Pt L3 edge, measured for positive (μ+) and negative (μ−) photon helicities, respectively. μ+ and μ− are normalized to unity such that the polarization-averaged x-ray absorption is unity, i.e. (μ+ + μ−)/2 = 1, at 11.567 keV before excitation. XMCD is defined as the difference of x-ray absorption of different helicities, Δμ = μ+ − μ−. The clear XMCD spectra with the magnitude of ∼15% with respect to the edge jump was observed and are in good agreement with the results obtained using a synchrotron beamline [44].

TrXMCD measurements were performed at an x-ray energy of 11.567 keV, at which the Pt XMCD spectrum reaches the maximum. Figure 2(b) shows the dependence of the trXMCD amplitude on the fluence of the pump laser at a fixed delay time of 100 ps. With the fluence of 40–80 mJ cm−2, the XMCD intensity is almost constant. Above 80 mJ cm−2, both μ+ and μ− decreased simultaneously, which was a signature of a destructive damage of the sample due to the strong irradiation by the optical laser. Therefore, we set the laser fluence to 32 mJ cm−2 to avoid damaging the sample and this condition was therefore used to obtain the trXMCD data presented in the following. With the laser fluence of 32 mJ cm−2, we repeated the delay scans and no significant change in the delay scan results was detected, excluding successive damaging of the sample. The results of pump–probe delay scan are shown in figure 2(c). X-ray absorption signals of μ+ and μ− varied symmetrically with the delay time.

Figure 1. Schematics of XFEL-based time-resolved XMCD measurements on FePt in the hard x-ray regime. (a) Experimental setup for pump–probe trXMCD constructed at SACLA BL3. (b) Schematics of capturing the ultrafast transient dynamics of the Pt moments. A fs optical excitation of L10-FePt thin films causes ultrafast dynamics of the Fe and Pt atomic moments. Ultrashort XFEL radiation pulses at the Pt L3 edge are subsequently employed to measure the dynamics of the Pt moments. (c) Fluorescence spectrum of Pt (3d→2p). The shaded area indicates the energy window of the Pt Lα emission line.
towards $\mu^+ = \mu^- = 1$. This behavior demonstrates fast demagnetization of the Pt 5d magnetic moment, which has been firstly observed by the element-specific hard x-ray trXMCD technique presently developed. The characteristic demagnetization time $\tau_{Pt}$ was calculated via fitting using a single exponential function:

$$f(t) = 0 \ (t \leq 0) \ \text{and} \ a[1 - \exp(-t/\tau_{Pt})] \ (t > 0)$$

convoluted by a Gaussian function to reflect the temporal resolution of 50 fs. We determined that the decay time is $\tau_{Pt} = 0.61 \pm 0.04$ ps. The fitting result is shown by the dashed lines in figure 2(c). At longer delay times shown in the figure 2(d), both $\mu^+$ and $\mu^-$ recovered to the values to those before irradiation of the optical pump laser within 1 ns. The time scale of recovery starting around 200 ps after slower recovery is estimated to be 260 ps via fitting the results using the exponential function.

To characterize the dynamics of the sample’s total magnetization, we performed trMOKE measurements for the same pump laser fluence as used in trXMCD. Figure 2(e) shows trMOKE results of L1$_0$-FePt for a pump laser fluence of 32 mJ cm$^{-2}$. The Kerr rotation angle $\theta_{MOKE}$ normalized by the angle before excitation $\theta_{MOKE,0}$ reduces to about 40% of that in the unexcited state. The time scale of demagnetization is estimated to be $\tau_{total} < 0.1$ ps via fitting with a single exponential function as in the case of trXMCD. The time constant $\tau_{total}$ is comparable to that in the previous trMOKE study, i.e. 0.15–0.38 ps [45] and to that of 0.16 ps measured for the pristine Fe case [46]. Figure 2(f) shows the trMOKE and trXMCD demagnetization and magnetization recovery for longer delay times. The time scale of recovery is estimated to be 200 ps from the trMOKE results shown in figure 2(f); this value is close to the recovery time scale of Pt magnetization, i.e. 260 ps, which has been determined from the trXMCD results. This similarity of the time scales indicates that the Pt and total magnetic moment nearly attain an equilibrium state during the recovery process in 200–260 ps. We further note that the Pt-trXMCD signal shows a stronger demagnetization and follows in its recovery after the trMOKE signal.

The ultrafast demagnetization dynamics obtained with trXMCD and trMOKE are plotted together in figure 3(b) for $t < 4$ ps. The XMCD amplitudes and Kerr rotation angles have been normalized to the values of the unexcited state ($t < 0$) to discuss the relative changes in the Pt magnetic moment and in the total magnetization. It is known that the Fe magnetic moment dominates the total magnetic moment. It responsible
for about 88% of the total moment [44]. To investigate the contribution of the Fe and Pt moments to the measured Kerr rotation, we have performed *ab initio* calculations of the MOKE spectrum [47], shown in figure 3(c), and the details are described in the appendix. In these calculations we find only a small reduction of the computed Kerr rotation when we set the magnetic moment on Pt to zero, but without Fe moment the signal totally vanishes. This substantiates that the trMOKE measurement reflects the Fe magnetization and can be used to discuss the ultrafast dynamics of the Fe magnetic moment. One can see in figure 3(b) that Fe demagnetizes much faster than Pt, which was characterized by the different demagnetization times, $\tau_{\text{total}} < 0.1$ ps and $\tau_{\text{Pt}} = 0.6$ ps. However, with regard to the amplitude of demagnetization, Pt becomes more demagnetized than Fe (at $t > 1$ ps). The Pt magnetic moment decreases to 20% of the full magnetized state, while the Fe magnetic moment retains more than 40% of the full value. This means that *transiently* the ratio of Fe and Pt magnetic moments changes. The absolute value of the Fe magnetic moment is still larger than that of Pt in the region of $0 < t < 0.6$ ps, where the Fe magnetization decreases very rapidly. The observed ultrafast modulation of the ratio of moments just after photo-excitation is consistent with previous measurements on GdFeCo [6]. But note that here we report this for an induced magnetic moment, and also, the changed ratio remains on a longer time scale of tens of ps which seems to be unique in our case.

3. Discussion

An important aspect to explain the element-dependent demagnetization timescales is the excitation depth of the pump laser and the probing depths of the trMOKE and trXMCD techniques. The 1.5 eV pump laser has an excitation depth of about 10 nm in the 20 nm FePt sample. Our trMOKE set-up employing 2 eV probe radiation probes a similar, 10 nm, sample depth. On the other hand, trXMCD measures the Pt response in the whole sample. This penetration depth mismatch is expected to increase $\tau_{\text{Pt}}$ because the time for the demagnetized area to expand is included in the observed $\tau_{\text{Pt}}$. A further important aspect is that Fe has a significant partial density of states (pDOS) below $E_F$ that can be excited by the laser pump, and even has a larger pDOS than Pt around $E_F$ [45, 48] because the Pt 5d bands lie at lower energies than the Fe 3d bands, which provide the main part of the DOS at $E_F$. This implies that spin-majority electrons of Fe 3d states will be excited to Pt states either immediately through optical intersite electron transfer [49] or by initial excitation to unoccupied Fe states followed by hot electron hopping to Pt sites as in superdiffusive transport [50]. The
latter is a semiclassical picture that captures the net changes in the local electron population stemming from modified extended wavefunctions. On the very short time during and immediately after the laser pump ($t = 0–0.1$ ps), effectively hot majority-spin electrons are thus transferred from Fe to Pt atoms in their vicinity (figure 3(a)). This causes a fast reduction of the Fe magnetization, but a slower reduction of the magnetization on Pt. During this initial nonequilibrium state the electrons scatter with one another and start to thermalize. The hot electrons will furthermore propagate through superdiffusive transport [50] from the laser-excited region deeper into the FePt film, similar to recent observations of superdiffusive spin transport in metallic heterostructures [12, 17, 51, 52]. The hot spin-polarized electrons transported into the deeper region are initially the more mobile majority-spin electrons [53]. The fast decay of the Fe moment measured with trMOKE can thus be explained by a rapid loss of spin polarization to neighboring Pt atoms and subsequent fast spin transport occurring within several hundreds of femtoseconds into the deeper sample region where these spins are no longer probed by trMOKE. The trXMCD conversely probes the Pt response of the pump-laser excited region and the deeper lying region, and will therefore in the first few hundred femtoseconds detect a smaller demagnetization than that measured with trMOKE. The process of superdiffusive transport seizes as the excited electrons scatter and thermalize in about 500 fs [54].

The hot electrons will also quickly start to transfer energy and angular momentum to other degrees of freedom such as lattice and spin excitations [5]. At about 1–2 ps the dominant part of this energy and spin redistribution in the sample is completed and incoherent transversal spin excitations [55] with a high spin temperature are present on the Fe atoms. The fact that the moment on Pt is induced by the ferromagnetic element Fe [56] is now important to understand why the normalized Pt moment at $t > 1$ ps is reduced more than that of Fe. In figure 3(d) we show the $ab$ initio calculated reduction of the Fe and Pt moments due to transversal spin motion with an average angle with respect to the $c$ axis, which is the easy axis. The Fe spin moment behaves essentially as a length-conserving Heisenberg moment, but the Pt moment behaves differently. Assuming, for example, an average $c$ axis angle of 60° gives an average $z$ component of the normalized Fe moment $M_z / M_0$ of 0.5. However, due to the induced nature, the $z$ component of the normalized Pt moment is much more reduced, to $\sim 0.3$, consistent with the measured ratio of Fe and Pt spin moments at about 2 ps. Thus, the observed stronger reduction of the moment on Pt is a consequence of the induced nature of the Pt moment in combination with excitation of high-amplitude incoherent transversal spin oscillations. For this transient magnetization moment around $t \approx 3$ ps the penetration depth effect is considered to be limited. The demagnetization is spread to the whole film, and the demagnetization observed by trXMCD is larger than that of trMOKE. This implies that a transient magnetization ratio modulation exists around $t \approx 3$ ps.

Since FePt is considered as one of the most promising materials for high-density magnetic recording, its demagnetization behavior under laser-excitation has drawn attention recently [3, 45, 57]. The element-dependent demagnetization of CoPt, NiPd, and FePt alloys have been studied recently with trMOKE in the EUV regime (energies of 50–70 eV) [19–21]. Hofherr et al. [21] observed for FePt similar demagnetization times on both Fe and Pt, in contrast to our measurements that show a disproportionate demagnetization. The origin of the different demagnetization behaviors requires further investigations. We note that the closeness of the Fe M edge (54 eV) and Pt O$_2$ edge (65 eV) and O$_3$ (52 eV) energies causes a stronger mixing of Fe and Pt signals in the EUV trMOKE. It is however evident that our hard x-ray trXMCD measures compellingly the atomic Pt 5d demagnetization dynamics. In addition, we note that the magnetic anisotropy is different in the investigation of Hofherr et al; in their case the sample is polycrystalline and has the chemically disordered fcc structure whereas our sample is magnetized in the out-of-plane direction.

4. Conclusions

We have studied the Pt magnetization dynamics in L1$_0$-FePt thin films with trXMCD using ultrashort and circularly polarized XFEL pulses at the Pt L edge in the hard x-ray regime. With the complementary use of trMOKE measurements we demonstrated an element-specific demagnetization process with different decay times of Pt and the total, Fe-dominated, magnetization as $\tau_{\text{Pt}} = 0.6$ ps and $\tau_{\text{total}} < 0.1$ ps, and evidenced a transient magnetic state after 1 ps, wherein the ratio of the magnetic moments of Pt and Fe was changed. We explained this distinct demagnetization behavior by the Fe-exchange induced character of the Pt moment, which reduces stronger when incoherent transversal spin excitations are present on the Fe sublattice. It should be noted, though, that our result is obtained on this one sample and that the laser fluence used was fixed to the specified value. The effect of laser fluence and dependence on the sample thickness should be clarified by further experimental and theoretical studies. Further trXMCD experiments on both the Fe and Pt edges need to be performed while changing the experimental conditions systematically. In the future, our newly-developed trXMCD technique can be extended to the Pt L$_2$ edge and transient spin and orbital magnetic moments could be obtained by sum rule analysis, which then could further be complemented by soft-x-ray trXMCD spin and
orbital moment measurements at the Fe L edges [58]. We therefore anticipate that high-resolution hard x-ray trXMCD measurements can shed new light on the still poorly understood atom-specific demagnetization dynamics of composite magnetic materials containing heavy elements.

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Appendix. Sample characterization and ab initio calculations

The single-crystal L10-FePt thin films with a thickness of 20 nm were epitaxially grown on MgO(100) substrates using the sputtering method at a substrate temperature of 500° C. Figure A1 shows the magnetic field dependence of MOKE angle, where the magnetic field was applied in the normal direction to the film plane. The measurement was performed at room temperature and the wavelength of the probing light was 680 nm.

To characterize further the dynamics of the sample’s magnetization, trMOKE measurements were performed using visible laser as pump and probe light. A Ti:sapphire regenerative amplifier with a photon energy of 1.5 eV, a pulse width of 100 fs, and a repetition rate of 1 kHz was used as the light source. In trMOKE experiment, pump and probe light comes from the same laser and we used an optical delay stage for generating time delays. The timing jitter was small enough that we did not need any jitter collection. The timing zero difference seen in figure 3(b) results from the time resolution difference. The experimental results and fitting curves plotted in figure 3(b) are shifted based on the timing zero determined by fitting. The pump beam was a fundamental light, and its repetition rate was decreased to the half of the fundamental frequency (500 Hz) with an optical chopper. The probe beam with a fundamental frequency of 1 kHz had a photon energy of 2 eV which converted by an optical parametric amplifier, and was incident nearly normal to the film surface, after which the polarization of the reflected light was analyzed.

For the electronic structure calculations we have adopted the relativistic density functional theory framework in the local spin-density approximation [59]. To compute the atom–resolved MOKE spectra we have employed the Kubo linear–response theory to evaluate the optical conductivity tensor elements on the basis of the selfconsistent Bloch wavefunctions and eigenenergies, using an in-house code based on the augmented spherical wave electronic structure method, see [47] for details. The contributions stemming from the Fe and Pt atom, respectively, to the MOKE spectrum were computed by using the selfconsistently converged FePt electronic structure in combination with a single-shot, non-selfconsistent MOKE calculation in which the influence of the zero Pt (or Fe) moment is included in the optical transition matrix elements of the optical conductivity tensor. To obtain the angle-dependent magnitude of the Fe and Pt spin and orbital moments the selfconsistent FePt electronic structure for noncollinear configurations of the Fe and Pt moments was calculated for each canting angle using the tight-binding linear muffin-tin orbital (TB–LMTO) method [60].

![Figure A1. Measured magnetic hysteresis loop of 20 nm FePt film.](image-url)
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