Multi-Color Imaging of Magnetic Co/Pt Multilayers

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We demonstrate for the first time the realization of a spatial resolved two color, element-specific imaging experiment at the free-electron laser facility FERMI. Coherent imaging using Fourier transform holography was used to achieve direct real space access to the nanometer length scale of magnetic domains of Co/Pt heterostructures via the element-specific magnetic dichroism in the extreme ultraviolet spectral range. As a first step to implement this technique for studies of ultrafast phenomena we present the spatially resolved response of magnetic domains upon femtosecond laser excitation.

Index Terms— **Co/Pt, demagnetization, free-electron lasers (FEL), holography, magnetic domains, magnetic multilayers, perpendicular magnetic anisotropy, X-ray magnetic circular dichroism (XMCD).**

I. Introduction

ULTRATHIN perpendicularly magnetized heterostructures, such as the two-compound multilayer system Co/Pt, show fascinating physical properties due to strong spin-orbit coupling [1]. Co/Pt multilayers have a significant interface contribution to the perpendicular magnetic anisotropy which can be used for the control of domain wall motion arising from an interplay of Dzyaloshinskii–Moriya interaction [2], [3] at the Co/Pt interfaces and spin currents [4]. Also based on these mechanisms room-temperature skyrmions can be created in thin Co/Pt films in racetrack memories [5]. Optical induced ultrafast spin currents in ferromagnetic thin-film structures pave the way for future ultrafast spintronic devices [6] and broadband terahertz emitters [7]. Moreover, Co/Pt thin-film magnetism has a huge potential in magnetic storage devices. Not only because of the fulfilled requirements of long-term stability due to the strong magnetic anisotropy but also because of recently discovered helicity-dependent all-optical control of ferromagnetic multilayers [8], [9]. However, many underlying microscopic mechanisms on ultrafast time and nanometer length scale are not fully understood and currently under debate. In particular, the origin of all-optical helicity-dependent switching or the fundamental microscopic processes leading to an optically induced ultrafast demagnetization on a femtosecond time the scale of ferromagnetic materials [10] raise many intriguing questions. To gain more insight into the interaction between non-local spin currents and interacting multi-compound perpendicularly magnetized heterostructures requires novel techniques that give a direct and simultaneous view of the element-specific magnetization on a femtosecond time and nanometer length scale.

Here, we present the first experimental demonstration of two-color imaging with Fourier transform holography (FTH) in the extreme ultraviolet (XUV) spectral range measured at the free-electron laser (FEL) facility FERMI in Trieste, Italy. The reconstructed magnetic domain pattern gives simultaneously element specific and real space access to the magnetization of a two-compound Co/Pt multilayer system. Furthermore, a time-resolved single-color approach reveals spatially resolved frames of the optical induced ultrafast demagnetization dynamics of Co/Pt. The feasibility and the prospect of combining both, multi-color spectroscopy with nanometer resolution and femtosecond spatial resolution are discussed.

II. Experimental Setup and Results

The coherent imaging experiment was performed at the FEL facility FERMI at the end-station DiProI [11], [12] combining femtosecond temporal and nanometer spatial resolution in an optical pump—XUV probe setup. To image the magnetic domains of a Co/Pt multilayer an FTH technique in transmission configuration [13]–[15] were used. In this imaging technique a hologram is formed by interference of an object with a known reference wave. In the XUV to soft X-ray spectral range this readily yields spatial resolutions on the order of approximately 30 to 80 nm. For optimal stability the sample and the reference holes are manufactured in a monolithic mask design. The 250 nm thick gold mask with no transmission in the XUV spectral range is evaporated on a 30 nm thin silicon nitride membrane supported by a 200 μm thick silicon substrate. A circular hole with a diameter of $d_r = 2$ μm is drilled in the mask via a focused ion beam and defines

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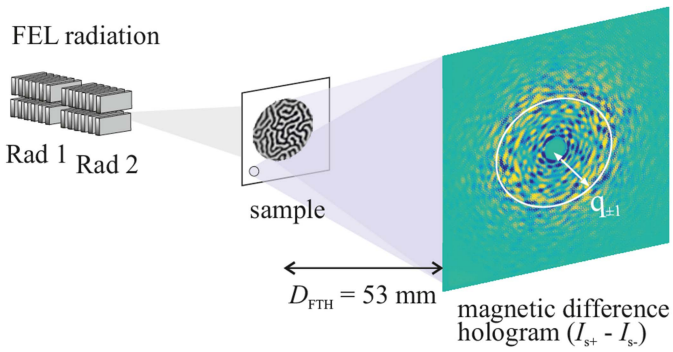

Fig. 1. Sketch of the FTH setup carried out at the FEL at FERMI. FEL radiation in the XUV spectral range is created in the undulators Rad 1 and 2, illuminating the Co/Pt multilayer that is structured with a holographic gold mask. The diffracted radiation centered at the Co M-edge at 60 eV originating from the reference wave and object wave interferes on a CCD detector placed 53 mm behind the sample. A beam stop cuts out the intense direct beam. The difference hologram between left and right circular polarization contains only magnetic information.

the object hole (field of view). Afterwards, the magnetic film Al(10)/Pt(2)/[Co(0.6)/Pt(0.8)]$_{20}$/Al(3) nm is deposited on the opposite side of the membrane via magnetron sputtering. Due to prior sample characterizations via magnetic force microscopy and small angle X-ray scattering the domain size is determined to be on the order of 80 nm. In the last step five reference holes with diameters ranging from 60 to 80 nm are drilled through the entire sample. Each reference will generate a twin-image of the object in the reconstruction leading to ten reference-object cross correlations. To increase the signal to noise ratio of the reconstruction, images stemming from the different reference holes can be added. To avoid an overlap of the reconstructed objects with the object–object or reference–reference auto correlation in the center of the reconstruction, all references were arranged in a circle of $|\vec{r}| = 5\ \mu$m diameter around the object hole, equidistantly placed with respect to each other.

The time-resolved single-color and static two-color coherent imaging experiment uses probe pulses in the XUV spectral range. The radiation is tuned to the Co M-edge and Pt N-edge for resonant small angle scattering due to the magnetic circular dichroism (MCD) effect. Magnetic sensitivity to Pt is confined to the Co/Pt interface where band hybridization induces magnetism in Pt. The scattered intensity of reference and object holes form the interference pattern (hologram), which is recorded on a charge-coupled device (CCD) detector placed 53 mm behind the sample (Fig. 1). The difference image between holograms recorded with left/right circularly polarized radiation yields the magnetic information. To increase the dynamic range of the recordable intensity and to avoid damage to the CCD detector a beam stop was used to block the intense direct beam.

The achievable spatial resolution is determined by the size of the reference hole and the maximum-recorded scattering vector $q_{\max}$ which is determined by

$$q_{\max} = \frac{4\pi}{\lambda} \sin\theta_{\max} \tag{1}$$

with the wavelength λ and the maximum scattering angle $\theta_{\max}$ that is given by the size of the CCD chip and the sample-CCD distance. The experimental setup enables a maximum recordable wave vector of $q_{\max} = 77\ \mu\text{m}^{-1}$ at 20.8 nm (Co M-edge) and $q_{\max} = 92\ \mu\text{m}^{-1}$ at 17.3 nm (Pt N-edge) leading to resolvable spatial frequencies of $d = 2\pi/q_{\max} =$ 82 and 68 nm, respectively. Also note that in the XUV spectral range wave guiding effects of the reference hole may further influence the spatial resolution.

As a significant number of photons with large scattering angles are required, we average several hundreds to thousands of FEL pulses to form the hologram. At the same time, the XUV intensity of the FEL pulses in the imaging experiments has to be reduced to prevent non-reversible changes and damage of the magnetic domain network [16], [17].

A. Time-Resolved Single-Color Imaging of Co/Pt

For the time-resolved single-color imaging of Co/Pt we optically excite with laser radiation centered at 800 nm. The laser-triggered ultrafast dynamics are probed by coherent XUV radiation at the M-edge of Co at 60 eV. The laser fluenc was set to approximately 3 mJ/cm^2. The pulselength of the optical pump beam and the XUV probe beam is on the order of 100 fs. For each helicity we integrated 10 min with a 10 Hz repetition rate, i.e., 6000 pulses.

In Fig. 2(a), we show the spatially resolved reconstruction of the magnetic domains in an unpumped state 1 ps before time zero. The clearly resolved magnetic domains with a known periodicity of 160 nm allow a conservative estimation of the spatial resolution below 80 nm. Black and white areas represent out-of-plane worm-like domains pointing in and out of the surface while the surrounding gray area represents the border of the field of view. The cross section of the domain contrast (line) is displayed in more detail in Fig. 3(b). In Fig. 2(b), one can see the reconstructed domains within the very same field of view but in an excited state 1 ps after time zero. As the contrast settings of both images are scaled in the same way a direct comparison between the pumped and unpumped domain pattern is possible. The excited Co/Pt film shows a strongly reduced magnetic domain contrast caused by ultrafast demagnetization of the up and down domains. This is further emphasized by the direct comparison of the cross sections of the unpumped and pumped contrast, shown in solid and dashed-dotted lines, respectively, in Fig. 3(b). To investigate whether the magnetic domain pattern shows any non-reversible changes either due to the FEL probe or the optical excitation pulses, we subtract the scaled pumped reconstruction from the unpumped reconstruction. The scaling factor is determined with a spatially resolved minimization algorithm and yields 2.4, corresponding to a global demagnetization rate of ~42%. The result is depicted in Fig. 3(a) and shows an almost perfect difference image and no indication of large magnetic domain reconfiguration or non-uniform demagnetization. Due to the high spatial resolution and the high sensitivity for the magnetization, it is possible to identify and localize small changes of the domain network and, if required, take them into consideration in the data evaluation. Careful inspection of Fig. 3(a) suggests that we can indeed identify small areas of non-reversible changes, see black and white dots in Fig. 3(a). This analysis is expected

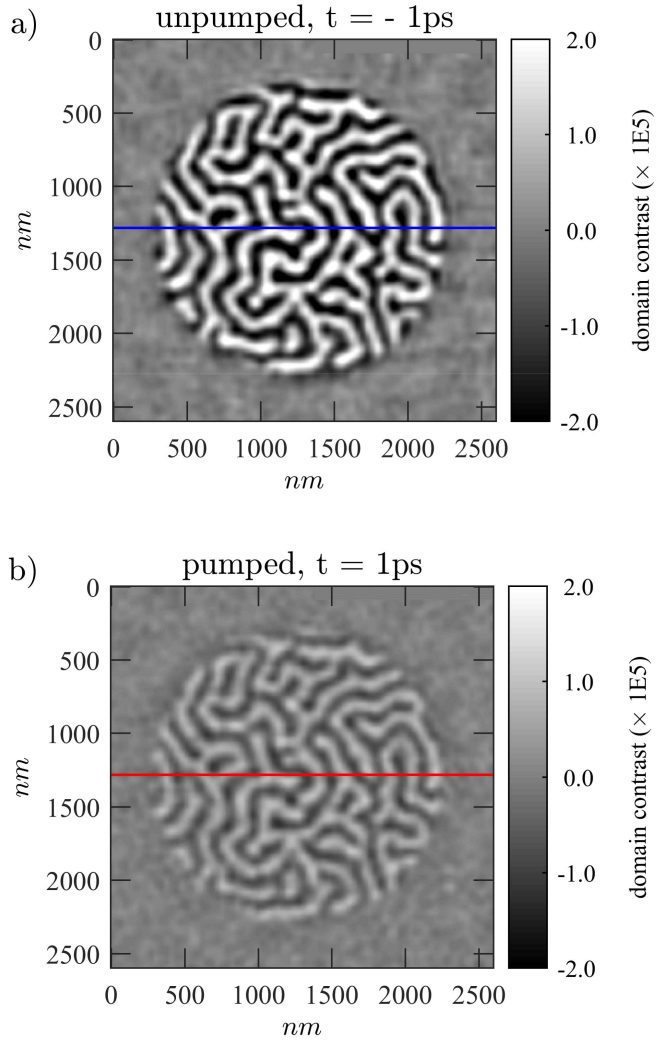

Fig. 2. (a) Difference holograms of an unpumped and (b) pumped magnetic domain pattern can be reconstructed with the help of a Fourier transformation. The result is visible in the two top panels showing a worm-like domain network within a field of view of 2 μm diameter.

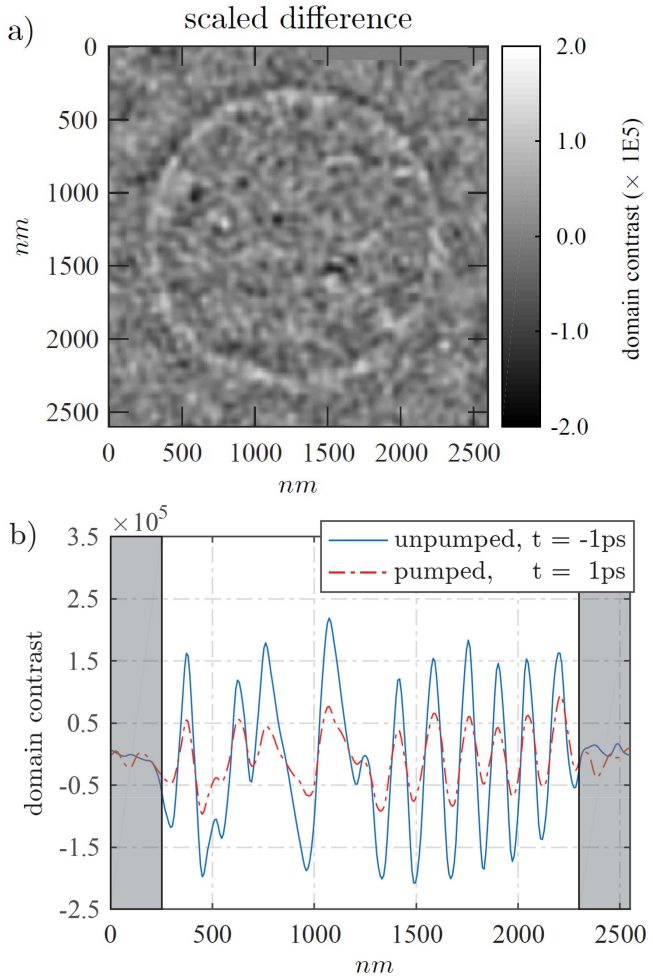

Fig. 3. Scaled difference of both reconstructions in (a) yields spatially resolved information about optical induced permanent changes of the domain structure. To compare the magnetic domain contrast of the Co/Pt multilayer between the excited (dashed-dotted lines) and not excited (solid line) state (b), two lineouts are plotted over the spatial coordinates. They not only show a stable domain position but also indicate that the amplitude of magnetization is quenched by approximately 40%.

to be particularly powerful in nano-structured samples where multiple reflections and interference of the incident pump light with the subwavelength dimension of the sample leads to an inhomogeneous intensity distribution. Controlling such complex light-matter interaction in nanostructures may allow to induce tailored absorption for nanoscale confinement of optically induced magnetization dynamics [12], [18].

Time resolved single-color imaging of Co/Pt using the MCD contrast mechanism is a sensitive and powerful tool for imaging magnetic heterostructures on a nanometer length and ultrafast time scale without inducing permanent changes or damaging their magnetic properties. That makes it the ideal tool for repetitive investigations of domain wall motion, skyrmion dynamics, and helicity-dependent all-optical switching in real space.

B. Static Multi-Color Imaging of Co/Pt

For simultaneous probing of the element-specific response of the magnetization of the Co film and the Co/Pt interface, we performed a static two-color imaging. This is possible due to the recent achievement at FERMI to produce two-color FEL radiation at integer multiples of the UV seed laser [19].

An experiment at the synchrotron facility BESSY II revealed pronounced dichroic signals in the XUV spectral range stemming from Co and the Co/Pt interface [20]. MCD of comparable amplitude was found at 61.4 eV (20.2 nm) slightly off resonant at the Co $M_{2,3}$ edge and at 71.6 eV (17.3 nm) at the Pt N_7 edge. These FEL energies can be achieved by seeding the FEL with UV light at 242.2 nm and tuning the first part of the undulator, Rad1, to the 12th harmonic to probe Co and the second part of the undulator, Rad 2, to the 14th harmonic to probe Pt.

For the physical dimensions of the holography mask containing an object hole with a diameter of $d_r = 2$ μm and an object-reference distance of $|\vec{r}| = 5$ μm, one expects an overlap of the two reconstructions for the two respective wavelengths of 17.3 and 20.2 nm. To ensure that both reconstructions pertaining to Co and Pt do not spatially overlap, one can either reduce the size of the field of view along the dispersion direction $\vec{r}$ or increase the distance between object

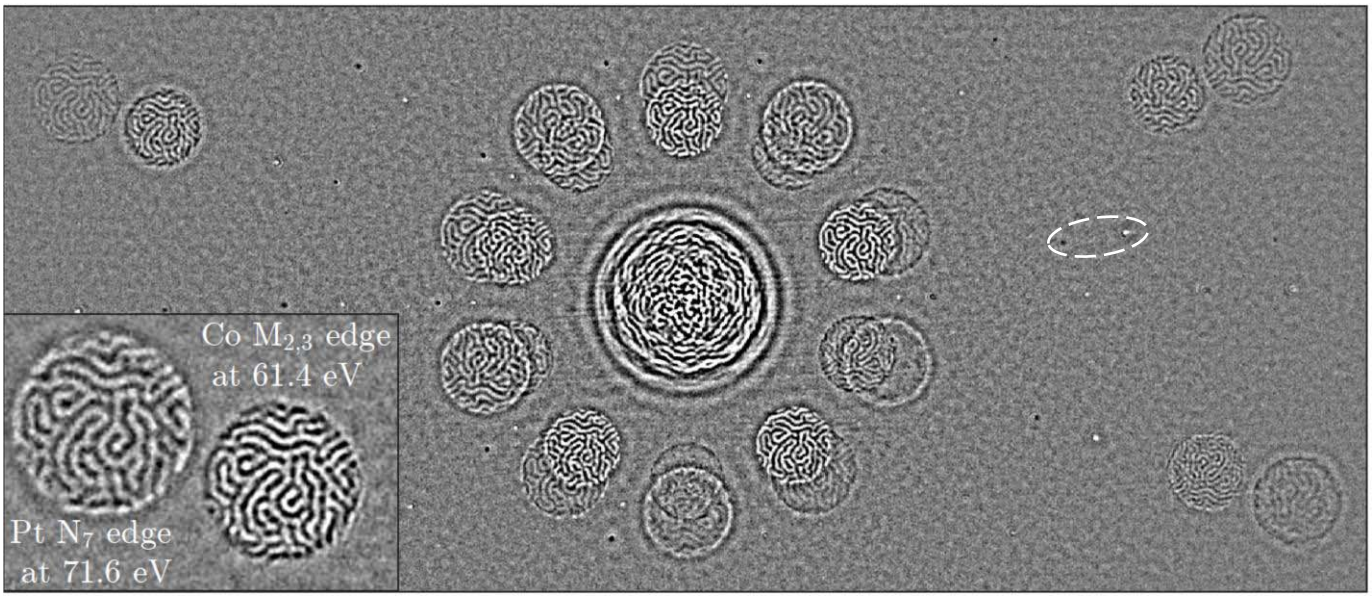

Fig. 4. Reconstruction of the static multi-color hologram of Co/Pt. All auto correlations are overlapped in the middle of the image. They are surrounded by a ring of 20 reconstructions of the object originating from the cross correlations between object and the five references for the two distinct energies. The two additionally references with $|\vec{r}| = 13$ μm lead to four clearly separated reconstructions of the magnetic domains coming from Co and the Co/Pt interface. For a more detailed depiction see the magnified inset. The small dots that are distributed over the reconstruction (see dashed white line for example) stemming from the cross correlations between the references sized between 60 and 80 nm.

and reference holes. More concretely, we have to fulfill the condition $|\vec{r}| > d_r\lambda/\Delta\lambda$. Since the FEL beam has a sufficient transverse coherence length, we add two additional reference holes at $|\vec{r}| = 13$ μm.

The energy of the two FEL pulses was set to 2 ± 0.3 μJ at 61.4 eV and 3 ± 0.3 μJ at 71.6 eV. With a spot size of 180 μm $\times$ 190 μm (full width at half maximum) this corresponds to 8×10^6 photons/μm^2/pulse and 10^7 photons/μm^2/pulse at the Co and Pt resonance, respectively. By adding up five images in 10 Hz mode with an integration time of 600 s for left and right helicity we obtain a hologram containing charge and magnetic contribution. A Fourier transformation of the difference hologram yields the direct visualization of a real space, element-specific and simultaneously recorded image of the magnetic domain network of Co and Pt with a high spatial resolution on the order of 80 nm.

The very first reconstruction of such a simultaneously probed two-compound system is shown in Fig. 4. It shows the Fourier transformation of a single difference hologram and reconstructs the magnetic domains from Co and from the Co/Pt interface. In the center of the reconstruction, all auto correlations (object–object and reference–reference) are located. They are surrounded by the cross correlations between object and reference holes. Note that we can also readily identify the cross correlations of the different reference holes showing up as white/black spots in the reconstruction. The fact that we can easily resolve them corroborates our conservative estimate of our spatial resolution on the order of 80 nm. As the position of the reconstructed object is energy dependent [21] we observe two displaced fields of view stemming from the two distinct XUV wavelengths. The well-separated cross correlations at the four edges originate from the two additional reference holes and the object. In the lower left corner, we show a magnified depiction of the element-specific reconstruction. Here we have increased the signal to noise ratio by summing up the two independent reconstructions from the two new reference holes. A careful comparison of the reconstructions from Co and from Pt yields an identical but inverted domain contrast. This is due to an opposite sign of the MCD effect of the Co M-edge (61.4 eV) and Pt N-edge (71.6 eV), i.e., the magnetization of the Co film and the Co/Pt interface point in the same direction [22].

Using this method in a dynamic manner can lead to a deeper understanding of the influence of localized Co/Pt interfaces on ultrafast demagnetization dynamics.

III. Conclusion

We demonstrated that single-color coherent imaging of Co/Pt heterostructures via FTH in the XUV spectral range is a powerful tool to gain detailed information about magnetization dynamics on the nanometer length and ultrafast time scale. Furthermore, we presented the very first implementation of a two-color imaging at the FEL facility FERMI by taking advantage of the MCD effect at the Co M-edge and the strong dichroic signal stemming from the Pt N-edge. It allows simultaneous and element-specific access to thin-film magnetism. By going from the M-edge to the L-edge of Co and Pt one could push the limit of spatial resolution below 20 nm which for example allows direct imaging of magnetic skyrmions.

We point out the feasibility of combining both approaches for setting up a time resolved, simultaneous and multi-element probing to unravel ultrafast processes in thin magnetic multi-compound heterostructures on a nanometer scale.

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