Magnetic imaging with soft x-ray spectroholography

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We present recent advances in imaging magnetic nanostructures via lensless holographic x-ray techniques. In addition to the tunable energy and polarization this method also uses the coherent photon flux that is now available at third generation synchrotron radiation sources. Our spectroscopic imaging technique is based on the direct Fourier inversion of a holographically formed interference pattern, thus we refer to it as spectroholography. By exploiting the magnetic dichroism in resonance at the $L_3$ absorption edges of the magnetic transition metals (wavelength of $\sim 1$–2 nm and energy of $\sim 700$–900 eV), images of magnetic nanostructures have been obtained with a spatial resolution below 50 nm. Using Si$_3$N$_4$-membrane substrates we combine in our experimental setup the sample itself rigidly with the holographic mask structure consisting of an 800 nm gold layer with a micronsized object aperture and a nanosized reference hole on the backside of the membrane. Such a compact implementation leaves plenty of space for installing extreme sample environments such as high magnetic fields and low temperatures that are difficult to combine with conventional x-ray imaging techniques based on focusing optics. © 2006 American Institute of Physics. [DOI: 10.1063/1.2165925]

Recently, we have demonstrated that spectroholography is a powerful x-ray technique for studying nanostructures. Besides scattering due to charge-density contrast one can exploit the resonant magnetic dichroism at, e.g., the $L_3$ edges of transition metals or the $M$ edges of rare-earth metals to image magnetic heterogeneity down to the nanometer length scale. By subtracting positive from negative helicity images it is possible to eliminate all charge scattering and extract the purely magnetic contrast. Our lensless technique is complementary to photoemission electron microscopy (PEEM) as well as transmission x-ray microscopy (TXM) and offers perspectives for spatial resolution down to the diffraction limit.

Here, we report on the current effort to develop spectroholography for the advanced characterization of magnetic nanostructures and recording media. Similar studies were performed in the past using incoherent resonant soft x-ray scattering techniques. However, spectroholography extends past studies towards real space imaging with high resolution and the potential of accessing ultra-short-time scales at future Free-electron laser (FEL) facilities.

In conventional coherent scattering experiments the field of view is defined by an aperture with diameter $\delta_o$ smaller than the transverse coherence length $\xi$ of the radiation used, or, in the case of an isolated small object, by the size of the object itself. In this case the recovery of the phase information remains difficult. Using a second, smaller reference pinhole with diameter $\delta_r$ at a distance $d$ from the object pinhole with $d > 3/2\delta_o + \delta_r$, and $(d + \delta_o + \delta_r) < \xi$, introduces a Fourier holography geometry that naturally preserves the relative phase between the larger object aperture and the smaller reference pinhole in the jointly formed interference pattern. Our basic setup is illustrated in Fig. 1. A coherent circular-polarized soft x-ray beam illuminates the sample/membrane/mask structure. The x rays scattered by the magnetic structure within the object aperture interfere with the undisturbed x-ray beam transmitted through the reference pinhole and form a holographic interference pattern on the charge-coupled device (CCD) detector that encodes the image of the magnetic nanostructures. This image can be reconstructed via a direct Fourier inversion using a single fast Fourier transformation (arrow in Fig. 2) with a computer as shown in the upper right inset. Due to the off-axis geometry in Fourier transform holography the object image and its conjugate can easily be separated (via the condition $d > 3/2\delta_o + \delta_r$). A more detailed description of the initial demonstration experiment can be found in Ref. 1.

Working in transmission geometry our experimental setup integrates the holographic mask structure, comprised by the object aperture and reference pinhole, directly to the...
sample. The sample is deposited on the front side of an x-ray transparent Si$_3$N$_4$ membrane substrate while an 800 nm x-ray opaque gold layer is deposited on the backside of the membrane. The object aperture and the reference pinhole are patterned into the gold layer using focused ion beam (FIB) lithography. Further technical details regarding the gold mask layer and the aperture as well as pinhole fabrication will be described elsewhere.\textsuperscript{13}

With our setup only very minor modifications are necessary to upgrade an existing scattering setup for spectroholography experiments. Since the holographic mask structure is directly integrated with the sample, the experimental space around the sample, can be used conveniently to apply high magnetic fields or low temperatures while drift and vibrations are essentially eliminated.

As a demonstration, Fig. 2 displays a sequence of spectroholography images of magnetic stripe domains that have been obtained from a [Co(12 Å)/Pt(8 Å)]\texttimes50 multilayer with perpendicular anisotropy during an external magnetic field sweep using a standard scattering setup (the ALICE reflectometer\textsuperscript{14}) that supplies fields up to ±6 kOe. All holography measurements have been performed at the Co-$L_3$ edge (~780 eV) with circular polarization at beamline UE52-SGM of the German soft x-ray synchrotron facility, BESSY, in Berlin (with a photon flux of about 10$^{11}$ photons/sec/0.1 Å/dE). The magneto-optical kerr effect (MOKE) hysteresis loop in the center of Fig. 2 reveals that the saturation field of the multilayer is slightly above 6 kOe. Thus we could not saturate the sample completely but rather move on a minor hysteresis loop that extends close to saturation. Corresponding spectroholography images are shown around the loop. Starting close to positive saturation we observe isolated bubble domains that grow out into one-dimensional stripes (at about 3.6 kOe) and coalesce at remanence to the labyrinth stripe domain pattern that is characteristic for perpendicular anisotropy systems.\textsuperscript{6,15} Approaching negative saturation the labyrinth decays back into isolated stripes (at about −4.5 kOe) and transforms into a bubble phase before reaching complete saturation. The transition from the isolated stripes into the bubble phase is often hidden due to the vanishing moment of a mostly low bubble density.\textsuperscript{16} However, in the thicker multilayer shown here, we observe a clear transition from stripes to bubbles in the macroscopic hysteresis loop due to an exceptional high density of bubble domains. Figure 3 reveals a distinct change in the slope of the hysteresis curve just before saturation. First, when contracting stripes towards bubbles we observe a steep slope in the $M$ vs $H$ plot and the magnetization decays quickly. In a second step, the remaining bubbles are annihilated and the magnetization decay slows down resulting in a distinct kink in the hysteresis loop as marked by the boundary point in Fig. 3. Earlier studies have shown that it is very difficult to finally annihilate all bubble domains and that fields well above the apparent saturation field might be necessary to reach microscopically complete saturation.\textsuperscript{16}
Finally, the resolution is limited by the fabrication of the flux for even smaller mask structures with higher resolution. However, with improved coherence, the sample can be placed behind the focus in order to obtain sufficient spatial coherence. In order to test the potential of our technique with the current fabrication techniques should be explored to improve the reference signal. Thus further aperture concepts, designs, and fabrication techniques should be explored to improve the reference signal and the image resolution.

In summary spectroholography is an emerging photon-in/photon-out technique for magnetic nanostructure characterization that combines the general advantages of synchrotron radiation (such as element specificity, i.e., separation of different magnetic layers, tuneable polarization, and direct proportionality to the magnetization $M$) with the necessary experimental space to create exceptional sample environments (such as high external magnetic fields and extreme temperatures) in a simple as well as vibration- and drift-free setup. In the future we plan to establish a spectroholography setup with external fields in the range of ±7 T down to temperatures of about 2 K at BESSY in Berlin. This end station will enable the imaging studies of the reversal behavior and switching field distribution for very hard materials, magnetic nanostructures and complex recording media. With the advent of free electron lasers (FEL) spectroholography will open a pathway to extend such magnetic studies to real time dynamics that reveal the ultimate timescales for magnetic reading and writing processes. With a fully coherent beam from a FEL a single femtosecond x-ray pulse will provide enough photons to obtain an image that currently needs integration over billions of pulses. Only then it will be possible to take advantage of the full potential of spectroholography to study individual magnetic processes with nanometer resolution on an ultrafast time scale.

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**FIG. 4.** MOKE hysteresis loop of a $[\text{Co}(0.4 \text{ nm})/\text{Pd}(1 \text{ nm})] \times 15$ multilayer deposited at 20 mTorr argon pressure. The inset in the center exhibits a spectroholography image in the out-of-plane demagnetized state. For this sample the diameter of the object aperture, which defines the field of view, is 940 nm with the reference pinhole diameter slightly below 100 nm. Thus typical exposure times for this sample were about 10–15 min per image.

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The sample shown in Figs. 2 and 3 was magnetron sputtered at an argon pressure of 3 mTorr resulting in smooth layers with a low coercivity and a regular stripe domain structure. In order to test the potential of our technique with respect to lateral resolution and resolvable Co thickness we deposited a magnetically harder multilayer at higher argon deposition pressure (20 mTorr) (Ref. 10) with fewer repeats and thinner Co layers. Overall the amount of Co was reduced by a factor of 10 resulting in a 100 times lower scattering intensity ($I_{\text{scat}} \sim M^2$). Figure 4 shows the corresponding MOKE hysteresis loop with high remanence and large coercivity similar to real magnetic recording media. The spectroholography image in the center of the loop is the difference between positive and negative helicity images. The magnetic domains are clearly visible even though they are smaller and more irregular than before due to the formation of exchange-coupled magnetic grains under high-pressure deposition conditions. Note that the diameter of the object aperture was reduced by a factor of 1.6 as compared to the previous figures. Recently we have pushed our studies to even thinner multilayer films (down to 2 nm total Co thickness) that have been deposited on artificially nanostructured substrates with pattern sizes of about 100 nm.

For the future, we plan to increase our resolution further to below the currently state of the art of 20 nm that limit other synchrotron techniques like PEEM and TXM. Such an increase in resolution could be reached by decreasing the diameter of both object aperture as well as reference pinhole, while compensating the lower signal by improving the coherent flux of the x rays. Using a partially coherent beam we currently have to place the sample significantly behind the focus in order to obtain sufficient spatial coherence. However, with improved coherence, the sample can be moved towards the focus thus providing an increased x-ray flux for even smaller mask structures with higher resolution. Finally, the resolution is limited by the fabrication of the pinholes using FIB techniques. Current reference pinholes are tapered with an exit diameter of about 50 nm and an aspect ratio of about 10 [see the lower left scanning electron micrograph (SEM) inset of Fig. 1]. Smaller reference apertures will help to improve the resolution, but at the cost of reference signal. Thus further aperture concepts, designs, and fabrication techniques should be explored to improve the reference signal and the image resolution.

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18. Image resolution roughly scales with the radius of the reference pinhole and in order to obtain significant interference between imaging and reference pinhole the relative intensities cannot be too different. Thus it is necessary to scale both pinholes down jointly, such that the cross interference term remains dominant in the coherent scattering pattern.