Studies of nanomagnetism using synchrotron-based x-ray photoemission electron microscopy

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Abstract. As interest in magnetic devices has increased over the last 20 years, research into nanomagnetism has experienced a corresponding growth. Device applications from magnetic storage to magnetic logic have compelled interest in the influence of geometry and finite size on magnetism and magnetic excitations, in particular where the smallest dimensions reach the important magnetic interaction length scales. The dynamical behavior of nanoscale magnets is an especially important subset of research, as these phenomena are both critical for device physics and profoundly influenced by finite size. At the same time, nanoscale systems offer unique geometries to promote and study model systems, such as magnetic vortices, leading to new fundamental insights into magnetization dynamics. A wide array of experimental and computational techniques have been applied to these problems. Among these, imaging techniques that provide real space information on the magnetic order are particularly useful. X-ray microscopy offers several advantages over scanning probe or optical techniques, such as high spatial resolution, element specificity, and the possibility for high time resolution. Here, we review recent contributions using static and time resolved x-ray photoemission electron microscopy to nanomagnetism research.

PACS numbers: 75.78.Fg, 75.78.Jp

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1. Introduction

Nanomagnetism, a discipline of studying magnetic phenomena unique to structures with dimensions in the submicron scale, is a particularly exciting area of research due to its fundamental role in physics as well as its potential technological applications [1,2]. A great example is the discovery of the giant magnetoresistance (GMR) effect [3,4], an effect of spin-dependent scattering in a nanostructured layered composite. The discovery of the GMR effect, which was awarded the 2007 Nobel Prize for Physics [5], led the way to an explosion of interest in nanoscale magnetic systems that exploit it. What followed was the development of an extraordinary effort to understand the physics of spin in electronic materials and to develop new devices around spin-dependent transport effects. This effort, known as “spintronics”, envisions replacing standard charge electronics with faster, denser, lower-power versions based on control of spin carrier populations. Manipulation of spin in electronics would also make possible new functions not easily achievable with charge electronics. These include non-volatile memory and, in principle, logic devices that may be reconfigured or reprogrammed on the fly. While much of the research into nanomagnetism and Spintronics remains at a very fundamental level, it has resulted in at least 3,200 publications in the last ten years [6], and has had a major impact on the development of new materials, deposition and lithography techniques, and magnetic characterization techniques.

Essential to the advancement of nanomagnetism is the development and application of a remarkable number of direct, real space magnetic imaging methods [7], among which synchrotron-based soft x-ray magnetic microscopy is extremely powerful. Very high chemical and magnetic contrast is available in this x-ray energy range resulting from strong core level absorption resonances and associated x-ray magnetic circular dichroism (XMCD) effects [8,9]. Consequently, numerous imaging techniques using the XMCD effect have been developed, often with achievable spatial resolution from 15 to 100 nm. The use of synchrotron radiation as the source for these microscopes also confers time resolution limited by the x-ray source pulse length (typically 50-100 ps at most 3rd generation sources), when used in a pump-probe mode. In this article we focus primarily on x-ray photoemission electron microscopy (X-PEEM), in which the local secondary photoelectron yield is electrostatically imaged, yielding a full-field image with a spatial resolution limited usually by aberrations in the electron optics. There have been significant contributions to our understanding of both the static and dynamic properties of nanomagnets through magnetic imaging techniques such as X-PEEM.

In this article, the contributions to nanomagnetism research using synchrotron-based X-PEEM is reviewed with the emphasis on the...
role of magnetic imaging, in particular time-resolved X-PEEM imaging, in nanomagnetism. Following a brief review of the development of the nanomagnetism field over the previous 10-15 years (Section 2) and an introduction to the PEEM technique (Section 3), in section 4 several recent PEEM experiments in nanomagnetism are covered. Here we review experimental results touching on major aspects of nanomagnetism, including interfacial coupling, finite size and shape effects, and magnetization dynamics. In Section 5 we outline a few current developments to the state of the art optics and sources and their potential impact to magnetic imaging and nanomagnetism, followed by a summary in section 6. Several comprehensive reviews dedicated to specific topics on nanomagnetism and magnetic imaging are available in the literature: Bader [1], (opportunities in nanomagnetism); Prinz [10], and Wolf et al [11] (Spintronics); Chien et al [12] (magnetic vortices); Vaz et al. [13], (ultrathin film structures); Stamps [14], and Nogués and Schuller [15], (exchange bias); Johnson et al. [16], and Sander, [17] (magnetic anisotropy); Eerenstein et al [18], Ramesh et al [19] (multiferroics); Freeman and Choi, [7], (magnetic microscopy); Ade and Stoll, [21, and refs therein] (a comprehensive review of x-ray microscopy techniques); Schönhenese [22], Kinoschita [23] Locatelli and Bauer [24], and Imada et al, [25] (PEEM); Schönhenese and Elmers [26] (time-resolved PEEM).

2. Overview of Nanomagnetism

Research into nanomagnetism encompasses a wide array of materials and types of nanoscaled structures. The development of advanced nanotechnologies, from chemically-driven self assembly of nanoscaled particles to the top-down lithographic approaches, enables the manipulation of materials at the nanoscale to realize new functionality. Nanoscaled magnetic materials with one or more dimensions at the nanoscale (10⁻⁷⁻¹⁰⁻⁹ meters), can be loosely categorized according to their dimensionality. As shown in figure 1, examples of nanoscaled magnetic materials include 0-dimensional nanodots, 1-dimensional nanowires, 2-dimensional ultrathin films or multilayers, and more complex structures that could have a combination of these characteristics. Nanoscaled magnetic materials often exhibit new and enhanced properties over their bulk counterparts, so they not only offer ideal material systems for exploring fundamental questions in nanomagnetism, but also hold promise for applications in information storage and magnetic sensing.

![Figure 1. Examples of nanoscaled magnetic materials: (a) 0-dimensional nanodots, (b) 1-dimensional nanowires, (c) 2-dimensional ultrathin films or multilayers, and more complex structures, such as (d) multilayered nano-rings, (e) nanocomposite films with embedded nanopillars (courtesy of R Ramesh) [19], and (f) artificial spin ice system (courtesy of D Lacour and S El Moussaoui)](image)

The magnetic configuration of a nanoscaled magnetic material is the result of energy minimization in such a material system. In general, the free energy of a nanomagnet is given by

$$E_{\text{total}} = E_{\text{H}} + E_{\text{exch}} + E_{\text{demag}} + E_{\text{anis}},$$  \hspace{1cm} (1)

where $E_{\text{H}}$ is the Zeeman energy, $E_{\text{exch}}$ is the exchange energy, $E_{\text{demag}}$ is the demagnetizing energy, and $E_{\text{anis}}$ is the anisotropy energy. The Zeeman energy, $E_{\text{H}}$, is proportional to the external magnetic field [27]. The exchange energy is most commonly described by the Heisenberg model with

$$E_{\text{exch}} = -\sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j,$$  \hspace{1cm} (2)

where $J_{ij}$ is the exchange integral determined by the overlap of the electronic wavefunctions of the two atoms with angular momenta $\mathbf{S}_i$ and $\mathbf{S}_j$ [28]. In ferromagnets (FM), $J_{ij}$ is predominantly positive, and thus favors a parallel alignment of magnetic moments, resulting in ferromagnetic ordering. In antiferromagnets (AFM), $J_{ij}$ is negative and favors an antiparallel alignment of magnetic moments.
The demagnetizing energy, \( E_{\text{demag}} \), is associated with the long-range dipolar interaction, favoring the formation of magnetic flux closure domains. The anisotropy energy, \( E_{\text{anis}} \), is due to magnetic anisotropies including magnetocrystalline anisotropy and surface anisotropy \cite{29}, favoring magnetization along certain crystallographic or surface directions.

When the size of the structure is comparable to the important length scales of these energies, the finite size will begin to have an influence on the spin configuration. The interplay between size confinement and proximity effect can result in unique novel magnetic configurations, such as magnetic domains and magnetic vortices, and interesting magnetic phenomena, such as the GMR effect, exchange bias and spin reorientation. Although some of the novel nanomagnetic effects have already been widely used in commercially available magnetic data storage devices, such as the exchange bias effect, many fundamental questions related to these phenomena have yet to be answered. The development and application of a remarkable number of direct, real space magnetic imaging methods, especially the synchrotron based x-ray photoemission electron microscopy (X-PEEM), make it possible to experimentally investigate these problems. Below we list some of such questions that have been extensively investigated by the X-PEEM technique.

2.1. Vortex dynamics in quasi-0D nanodots

When the sizes of all the three dimensions are shrunk to submicron scale, a quasi-0D nanodot is obtained. This type of structure is also often called a nanodisk when it is in a shape of flat disk. The magnetic configuration of a soft magnetic nanodisk is determined by the competition between the exchange and demagnetizing energies, because the magnetic anisotropy can be neglected in soft magnetic materials, such as FeNi alloys.

The ground state of a finite-sized magnet will typically try to minimize the field external to the magnet by dividing into domains, in order to reduce the demagnetizing energy. This can be accomplished if the local spins are oriented in random directions, such that the net moment of the structure is zero. Deviations from parallel alignment of spins costs exchange energy, however, thus the spins will only be randomized at length scales significantly above that of the exchange length. The result is that the magnetic will form into domains of local ferromagnetic order, with long-range randomization of the magnetization directions. These domains will have a characteristic size that is governed by both the exchange interaction and the magnetostatic coupling. When the size of the structure approaches this characteristic length, the finite size will begin to induce order. The consequence of this at the nanoscale is that the magnetization at the edges will try to align along the tangential directions. This will tend to drive the system to flux closure states. For a square structure, the simplest flux closure state is a four-domain state, with the magnetization directions at right angles to each other. Increasing the symmetry to a circular structure results in a continuous circulation of the magnetic flux around the disk. In this configuration, the spin directions are governed over most of the disk by dipolar interactions, except at the center, where the shorter-range exchange interaction dominates. In this region, approximately the size of the exchange length, the antiparallel alignment of spins would cost too much energy, so the system compromises by canting out-of-plane. This vertical core and circulating disk structure is a magnetic vortex state, as illustrated in figure 2. The magnetic vortex state is one of the most interesting and important nanomagnetic states.

Figure 2. Magnetic vortex state (courtesy of K Buchanan)

The vortex state is of interest for both fundamental and practical reasons. The unique topology of the vortex state allows the Landau-Lifshitz-Gilbert equation to be solved analytically, yielding great insights into magnetization dynamics at the fundamental level. They also have considerable importance for many device applications. Vortex states arise frequently in
magnetic domain walls, and can have a strong influence on their motion. Therefore, understanding vortex dynamics is very important for magnetization reversal, wall motion in nanowires, and switching of nanomagnetic devices. Also, because the magnetic vortex is a uniquely stable flux-closure state that displays two energy-equivalent polarities, there is interest in using them in non-volatile logic applications such as magnetic memories. Finally, the vortex state exhibits a well-defined eigenmode structure that offers the possibility of easily tuning through size, shape, or possibly external fields. This makes them very attractive for microwave applications.

The dynamics of the vortex state was considered theoretically first by Thiele [30] for the general case, and more recently in confined geometries by others [31, 32, 33, 34, 35]. These studies examined the excitation mode structure, and described the radial, azimuthal, and the vortex core translational modes. For a circular geometry, the Landau-Lifshitz-Gilbert equation can be written in the simplified form of a two-dimensional single particle equation of motion in the core position, \( \mathbf{X} \) (Thiele’s equation),

\[
M \dddot{\mathbf{X}} - \mathbf{G} \times \ddot{\mathbf{X}} + \frac{\partial W(\mathbf{X})}{\partial \mathbf{X}} = 0,
\]

(3)

This equation includes a cross term analogous to the Magnus Force in the motion of fluid vortices, such that the fundamental, or gyrotropic solution to this equation describes a circular trajectory of the core about the center of the disk, where the sense of the rotation is determined solely by the core polarity. The practical consequences in the presence of damping for a vortex core perturbed from the center are that it should follow a spiral path towards the center of the disk. Experimentally, the vortex core motion has been studied in both the frequency domain[32, 33Error! Bookmark not defined.], and with real-time magnetic imaging studies[36].

2.2. Domain wall motion in quasi-1D nanowires

While vortices offer the potential for using the core polarity as a non-volatile logic building block, there are considerable practical questions associated with addressing and sensing the state of such devices. Therefore, there is still considerable interest in using lower-symmetry magnetic structures for memory and sensing elements. In these more complicated geometries, finite size effects have a significant impact on magnetization reversal and the motion of magnetic domain walls. One specialized situation of interest is the magnetic nanowire, a quasi-1D geometry in which domain walls are inhibited in one direction by the small lateral size of the structure, but may form in the orthogonal direction. In principle, this allows more controlled manipulation of the domain wall, and magnetic logic devices [37, 38] and memory devices [39] have even been proposed based on based on the field-driven or current-driven domain wall motions, such as the domain-wall race track memory, as illustrated in figure 3 [39].

Magnetic field-driven domain wall motion has been extensively studied [40, 41, 42, 43]. In a nanowire geometry, Beach et al. [40] showed that for field-driven wall motion, the domain wall velocity is linearly dependent on the driving field, up to a limit of \( \sim 3-4 \) Oe. Above this limit, there is evidence for a more disordered motion. Micromagnetic simulations suggest that in this non-linear regime, the wall structure is modified such that a vortex forms [44, 45]. In this case, the motion of the vortex core is influenced by the same cross term as in the simpler disk geometry.
This leads to a non-monotonic domain wall position, and reversals of the core polarity when it encounters the edges of the wire [46, 47]. The result is a complex oscillatory motion of the wall position.

Current-induced domain wall has also been of great interests not only because of the promising applications in novel spintronics devices, but also because of the fundamental physics concerning the mutual interaction of magnetization and electric current [37, 48, 49, 50]. Although the prediction of the effect of electric current on a domain wall dated back to the 1970s [51, 52], the first experimental studies were reported much later [53] because the high-current densities required for driving domain wall motion was recently realized in lithography patterned magnetic nanowires. It is now generally accepted that the current-driven domain wall dynamics can be described by the Landau-Lifshitz-Gilbert equation with two additional adiabatic and nonadiabatic spin-transfer torque terms [54].

2.3. Magnetic coupling in quasi-2D multilayers

The magnetic multilayer is an interesting and rich system of both scientific and technological importance. In magnetic multilayers, different kinds of ultrathin layers are stacked sequentially in a periodic fashion. The advent of new film growth techniques makes it possible to fabricate intricate magnetic multilayer structures from various materials with thicknesses down to one atomic layer. The possibility of adjusting the constituent materials, layer thickness, and periodicity in the multilayers gives one the opportunity to fabricate multilayers with unique magnetic and transport properties. For example, magnetic multilayers consisting of alternating ferromagnetic and nonmagnetic metallic layers, such as Fe/Cr and Co/Cu, exhibit the GMR effect [3, 4, 55, 56]. In addition, the magnitude of the GMR oscillates as a function of the thickness of the nonmagnetic spacer layers, resulting from an oscillation in the sign of the interlayer exchange coupling between the ferromagnetic layers [57, 58]. Below we briefly review some interesting phenomena in multilayers investigated by X-PEEM technique, including exchange bias in ferromagnet / antiferromagnet hybrid structures, spin reorientation transition in ferromagnet / ferromagnet multilayers, and electric-field control of ferromagnetism in ferromagnet / multiferroic heterostructures.

2.3.1. Exchange bias in ferromagnet / antiferromagnet hybrid structures

Exchange bias, discovered by Meiklejohn and Bean in 1956 [59], is a very useful phenomena occurring in bilayers or multilayers of ferromagnet (FM) / antiferromagnet (AF) hybrid structures. Exchange bias refers to a shift in the soft magnetization hysteresis loop of the FM layer caused by the hard magnetization behavior of the adjacent AF layer, as illustrated in figure 4.

![Figure 4. The exchange bias phenomenon: easy-axis magnetization hysteresis loop of a soft ferromagnetic film (a) and an exchange-biased bilayer consisting of a ferromagnet and an antiferromagnet (b).]
equivalent, and instead there is one preferred easy magnetization direction for the FM, which is opposite to the direction of \( H_b \). The physics underlying this exchange bias phenomenon is the interfacial exchange interaction between the AF and FM. The FM layer is pinned by the AFM layer, and the magnetization reversal of the FM requires an added energetic cost, corresponding to the bias field \( H_b \).

The exchange bias phenomenon has been widely used in magnetic recording. The first commercial device to employ the exchange bias was IBM's anisotropic magnetoresistance (AMR) disk drive recording head. In the mid-1990s, an exchange bias layer started being used in the GMR based spin valve head. However, the origin of exchange bias phenomenon is still not well understood because of the difficulty in spatially resolving the magnetic domain structures in the AF layer and at the FM/AF interface as well as the correlation between the magnetic structure in the FM layer and that in the AF layer. A directly magnetic imaging technique with the sensitivity to magnetic configurations in AF films and the ability to resolve magnetic configurations at different layers is needed to better understand the origin of the exchange bias problem [60].

2.3.2. Magnetic anisotropy and spin reorientation transition in ultrathin magnetic multilayers

In ultrathin magnetic films or multilayers containing ultrathin magnetic layers, the complex interplay among the short-range exchange interaction, the long-range dipolar interaction, and the local magnetocrystalline anisotropy can cause magnetic anisotropy and domain formation to differ drastically from that in bulk magnetic systems [61, 62, 63].

Magnetic anisotropy determines the easy directions of magnetization in a sample and affects the process of magnetization reversal. Magnetic anisotropy of a magnetic sample is determined by the competition of the demagnetizing energy and the magnetic anisotropy energy, where the magnetic anisotropy energy depends on the magnetocrystalline anisotropy energy, magnetoelastic anisotropy energy, and the Néel surface anisotropy energy [16, 29]. The magnetocrystalline anisotropy is a relativistic manifestation of the coupling between the electron spin and the orbital moment, as first calculated by Bruno [64] and later experimentally verified by XMCD studies [65, 66, 67]. Therefore, modifications in the electronic structure at surfaces and interfaces causing a change of the spin-orbit coupling are expected to change the magnetic anisotropy. The magnetoelastic anisotropy depends on the strain in the film and the Néel surface anisotropy is related to the break of symmetry at the surface or interface. Both of the magnetoelastic anisotropy and the Néel surface anisotropy depend on the magnetic layer thickness. Therefore, magnetic anisotropy in epitaxial thin films or magnetic multilayers depends intricately on the film thickness. For example, figure 5 illustrates the thickness dependence of magnetic anisotropy in Co/Pd multilayers [16, 68].

![Figure 5. The thickness dependence of magnetic anisotropy in Co/Pd multilayers.](image)

The phenomenon, observed in many ultrathin films or magnetic multilayers, that the magnetization easy axis switches between out-of-plane and in-plane with magnetic film thickness is called spin reorientation transition (SRT) [69, 70]. Both magnetic materials with magnetization easy axis in the film plane and those with magnetization perpendicular to the film plane have important technological applications. Therefore, investigation and control of the magnetic anisotropy in magnetic multilayers are of both fundamental and application importance.
There are two main challenges for the experimental study of the SRT phenomenon. The first is that the possibility for sample-to-sample variations may overwhelm the effect to be studied. With the advancement of thin film deposition technology enables not only the film deposition precisely controlled at atomic layer thickness, but also the deposition of a wedged film, where the thickness of one layer is varied continuously in one single sample [69]. This wedge deposition technique greatly increases the uniformity of fabrication conditions, providing the solution to the sample-to-sample variations problem. This technique can also increase the effective throughput in both the sample fabrication and characterization steps.

The second challenge is the lack of experimental information on magnetic anisotropy and magnetic configuration of each individual magnetic layer in a multilayer system. Nowadays, the most widely used magnetic imaging method that can provide sufficient spatial resolution for the SRT study is magnetic force microscopy (MFM). In MFM, an image of magnetic contrast of the sample is obtained through the magnetostatic interaction between a ferromagnetic tip and the magnetic stray fields from the sample. However, the MFM imaging can neither provide the information on the origin of the magnetic anisotropy nor distinguish magnetic configuration of each individual layer in a multilayered sample [71]. A directly magnetic imaging technique with the sensitivity to magnetic anisotropy as well as the ability to detect magnetic configuration in each individual layer is needed.

2.3.3. Electric field control of ferromagnetism in ferromagnet / multiferroic heterostructures

Recently, an epitaxial bilayer of ferromagnet / multiferroic heterostructure has been experimentally studied by Chu et al, demonstrating the control of local ferromagnetism by electric field [72]. This work has added to the richness of the field of multilayers involving 2D magnetic layers and opened a new route for magnetism control in multilayered materials.

A ferroic is a material that possesses a spontaneous, stable, and switchable internal alignment [18]. For example, in ferromagnetic materials a spontaneous magnetization is stable and can be switched hysteretically by an applied magnetic field; in ferroelectric materials a spontaneous polarization is stable and can be switched hysteretically by an applied electric field; in ferroelastic materials a spontaneous deformation is stable and can be switched hysteretically by an applied stress. In general, a multiferroic is a material that exhibits more than one ferroic orderings simultaneously. However, in current convention, the term “multiferroic” primarily refers to a material that combines ferroelectricity with ferromagnetism, or more loosely, with any kind of magnetism [20]. Multiferroics have been of great interest because of not only the fundamental science but also their applications in magnetic data storage, spintronics and high-frequency magnetic devices [18-20, 73].

Although the pioneer works on multiferroics could be dated back to the 1960s and 1970s [74, 75, 76], the study of materials exhibiting coupled magnetic and electrical ordering has been revitalized in the last ten year or so because of both the advances in crystal synthesis, thin film deposition and various characterization techniques, and the improvement of first-principles computational techniques.

![Figure 6. Magnetic and ferroelectric coupling in ferromagnet / multiferroic heterostructures](image-url)
coupled with the multiferroic layer through the magnetic exchange coupling (FM) or exchange bias (AF). More importantly, the magnetization in the ferromagnetic layer can be controlled by an applied electric field through the coupling with ferroelectricity in the multiferroic layer.

In contrast to the extensive experimental studies reported on bulk or single phase thin film of multiferroics, relatively fewer results have been reported on the ferromagnet / multiferroic heterostructured materials because of the difficulties in not only sample fabrication but also heterostructure characterization. The X-PEEM technique provides a powerful tool for characterization of the interface coupling in such a heterostructure.

2.4. Magnetization reversal in complex nanostructures

In addition to 0-dimensional nanodots, 1-dimensional nanowires, 2-dimensional ultrathin films or multilayers, nanostructures can be more complex structures combining the above characteristics.

One example is a multilayer stack of alternating hard and soft ferromagnetic (FM) rings separated by a nonmagnetic Cu ring [77, 78] (as shown in figure 7), which is a very promising design for realizing a MRAM device [79, 80]. This pseudo-spin-valve design proposed by Zhu et al [77] intend to take advantage of the vertical GMR effect and the controlled chirality switching of the magnetic vortex states in each FM ring. The key point of this design is that the flux-closed vortex state reduces interactions between neighboring elements, enabling devices with high-density elements and robust and repeatable switching characteristics.

The second example is a nanocomposite thin film in which nanopillars of the FM phase embedded in a matrix of the multiferroic phase, as illustrated in figure 1 (e) [19]. Recently, epitaxial growth of ferromagnetic CoFe$_2$O$_4$ (CFO) nanopillars embedded in a single crystalline ferroelectric BaTiO$_3$ or BiFeO$_3$ (BFO) matrix has been demonstrated and an electric-field-induced magnetization switching has been observed in the BFO-CFO nanostructures [81, 82, 83].

The third example is an artificial spin ice system consisting of magnetic nanowires in a 2D lattice, as shown in figure 1 (f) [84, 85, 86, 87, 88]. In such an artificial spin ice system, each individual magnetic nanowire is a single-domain magnetic dipole. The interactions among the magnetic wires result in interesting phenomena, such as magnetic frustration [84, 89] and magnetic monopole effect [90, 91].

It is of great interest and significance to investigate the magnetic property of each individual component of the complex nanostructures, such as the soft and hard magnetic ring in the multilayered nanorings, and the ferromagnetic and multiferroic phases in the hybrid ferromagnetic-multiferroic nanocomposite films. The unique element-specific X-PEEM technique is an extremely powerful and appropriate tool for characterization of these complex nanostructures.

3. Photoemission electron microscopy (PEEM)

The basic principle of photoemission electron microscopy (PEEM) is to create a full field image of the electrons emitted from a sample surface in response to photon absorption. The focusing of the emitted electrons is usually done with a combination of electrostatic and/or magnetic lenses. Depending on the choice of excitation, this provides a surface-sensitive spatial map of some aspect of the samples electronic structure. Photoemission electron microscopy was first used with UV sources, and later came to exploit the greater element selectivity available by excitation with soft x-rays.

3.1. The instrumentation of PEEM

The structure of a basic electrostatic PEEM instrument is shown figure 7. The first extraction lens, along with the flat sample, forms an immersion-type objective. At the focal point of this objective, an aperture is usually placed to limit the angular acceptance of the system, and with that the spherical aberrations. This aperture is of variable size, thus an optimization can be achieved between total microscope transmission and spatial resolution. Following the aperture is a projection system usually consisting of two or three projection lenses. These form an image at the back focal plane, where an area detector is placed. The detector is most often a multichannel plate and scintillator screen, although solid-state detectors have been used in some applications. [92] Since
the detection scheme relies on photoelectrons, most of which are secondary electrons, the probing depth will be limited to approximately the electron mean free path in the sample. For soft x-ray excitation, the electron energies are typically less than 10eV therefore the escape depth is 2-5nm, according to the universal curve.[93]

**Figure 7.** Schematic diagram of an electrostatic PEEM instrument with x-ray excitation. Photoelectrons are stimulated by the x-ray beam, and are accelerated in to the column by the first extraction lens, which forms an immersion-type optic with the flat sample.

The performance, in terms of spatial resolution, of a PEEM is governed by three primary sources of focusing errors: spherical aberrations, chromatic aberrations, and limitations due to electron mean free path. Each of these may limit the achievable spatial resolution under certain operating conditions. For most electrostatic PEEMs, spherical aberrations limit the spatial resolution to ~20nm. However, with soft x-ray excitation, the kinetic energy distribution of the resulting photoelectrons is broadened, resulting in greater chromatic aberrations. In this case, the spatial resolution may be degraded to 50 to 100 nm. These errors may be minimized or corrected using additional optical elements. For chromatic aberrations, energy filtering of the photoelectrons using a retarding grid or electron spectrometer can improve the spatial resolution, at the cost of a reduced image intensity. Spherical aberrations can be corrected using electrostatic elements to compensate them. In Section 5, we discuss aberration-corrected microscopes in more detail.

### 3.2 The chemical sensitivity of PEEM

The spatial map of the photon absorption coefficient provides information about the details of the local electronic structure at the surface of the sample. The nature of the information obtained depends on the excitation energy used. With UV radiation, the contrast obtained is mainly derived from differences in work function across the surface. This can give chemical sensitivity, especially between metallic and oxidized surfaces. With excitation in the soft x-ray range (200 – 2000 eV), the absorption coefficient of most solid materials is dominated by strong core level resonances. With higher photon energies and tunable synchrotron sources, elemental contrast can be obtained by choosing an appropriate core level resonance. This can also provide sensitivity to valence state for many materials, since the local density of states in the final state will be sensitive to chemical bonding.

### 3.3 The magnetic sensitivity of PEEM

By manipulating the polarization of the incoming radiation, we can achieve sensitivity to spin and orbital occupancies. With excitation in the soft x-ray range, the absorption coefficient of most solid materials is dominated by strong core level resonances. Many of these resonances involve transitions to the unfilled bands where the magnetism of the material originates. These resonances may be quite strong, especially for 3d and 4f final states.

With circularly polarized radiation, there is an additional dependence of the absorption coefficient on the radiation polarization, resulting in a strong x-ray magnetic circular dichroism (XMCD) effect. This effect occurs for ferromagnetic materials and arises from the spin-dependent density of states in the final state band. The first experimental confirmation of the XMCD effect was at the Fe K edge (1s–4p) [94], however transitions that probe directly the states where the ferromagnetic spins reside typically show much greater effects [95]. For transition metals the L_{2,3} (2p-3d) transitions offer the largest effects, while in rare earths, the M_{4,5} edges probe the f states directly. In figure 8, we show schematically the energy levels for an L (2p – 3d) resonance of a 3d
metal, along with example Mn L edge absorption (I+ + I-) and dichroism (I+ - I-) spectra from a Mn compound film. As the incident photon energy is scanned through the core level binding energies, electrons are excited to the conduction band, creating a core hole. This core hole de-excites quickly, resulting in either a fluorescent photon or electron emission through Auger processes. The absorption coefficient may be measured by monitoring the yield of fluorescence photons, electron or by measuring the transmitted beam directly. For transition metal L edge absorption, the majority of the de-excitation events are in the form of electron emission, thus making this a convenient way to collect absorption data.

Two primary resonances, L$_3$ and L$_2$, are observed from the spin-orbit split 2p$_{3/2}$ and 2p$_{1/2}$ core levels, respectively. At each of these edges, a significant difference in absorption coefficient can be seen between left circularly polarized (LCP) and right circularly polarized (RCP) radiation, showing the XMCD effect. For 3d transition metals and 4f rare earth elements, this XMCD effect can be as high as ~30%, which translates into very high sensitivity. At state of the art soft x-ray beamlines, detection of magnetic moments as small as 0.01 $\mu_B$/atom is possible [96], and even very small induced moments in magnetically-doped materials or at interfaces can often be detected [97]. X-ray absorption and magnetic circular dichroism is also very sensitive to the band structure in the final states, making it a very effective probe of the local electronic structure. Finally, quantitative information can be extracted about the spin and orbital moments through the application of sum rules derived by Thole and coworkers [98,99,100].

The sensitivity to spin has been used extensively for imaging of magnetic materials and structures. Using circularly polarized radiation, the XMCD effect that occurs at certain resonances can be exploited to create an image of the projection of the local magnetization along the beam propagation direction. In the ferromagnetic transition metals, magnetic moments originate from the exchange-split d-bands, thus the transitions of interest are the L$_{2,3}$ (2p-3d) absorption edges, which mostly occur between 500 and 1000 eV. Since in this energy range the absorption cross section is still quite high, and the XMCD effect is very large at the L edges, very high sensitivity magnetic images can be obtained. Similarly, for the rare earth elements, the magnetically important M$_{4,5}$ (3d-4f) edges fall in the 900 to 1500 eV energy range, making X-PEEM a very attractive method for magnetic imaging. Dichroic effects exist at other atomic transitions where magnetic moments may be present, such as the oxygen K, transition metal K, and L edges of semiconductors. In principle, these could be used for magnetic microscopy as well. For PEEM imaging, however, the flux intensive nature of the PEEM technique makes it difficult to obtain sufficient signal to noise for these transitions. Typically, a magnetic X-PEEM image is formed by collecting left- and right-circularly polarized images at the appropriate resonance and taking the difference divided by the sum, as is the practice for XMCD spectroscopy. It is also possible to obtain magnetic images using the
contrast between $L_3$ and $L_2$ instead. In the case of transition metals, this is accomplished by dividing an $L_3$ image by an $L_2$ image. This has the advantage of not requiring a polarization change, which can be time-consuming or difficult with some types of sources.

In addition to dichroism between circular polarizations, it is possible to observe spin-dependent effects with linearly polarized light. X-ray magnetic linear dichroism (XMLD) arises when the spin-orbit coupling creates a directionally dependent sub-orbital population. Then, orbitals oriented parallel and perpendicular to the electric field axis of the incident photon will exhibit differing absorption coefficients. This effect is more prominent in materials that exhibit strong multiplet splitting, and is usually manifested as a difference in peak height between two multiplets. Because the XMLD effect is only sensitive to the angle between the atomic moments and the average electric field vector, it exhibits a $\cos^2 \theta$ dependence. Therefore, while the spin direction cannot be determined, XMLD is sensitive to the spin axis and the magnitude of the atomic moments. Consequently, XMLD is useful for studying antiferromagnetic order. Because other, non-magnetic, effects can contribute to linear dichroism, care must be taken to account for them.

3.2 Time-resolved X-PEEM

Using X-PEEM at synchrotron sources conveys advantages beyond high flux, chemical and magnetic sensitivity. The pulsed nature of synchrotron radiation provides a natural ability to obtain temporal data with a time resolution related to the x-ray pulse width. This pulse width varies between facilities and operating modes, but it typically between 50 and 200 ps, thus offering high sensitivity magnetic imaging with very high time resolution. Therefore, time-resolved X-PEEM is uniquely suited to tackle many of the most compelling problems of magnetization dynamics in confined structures. The number of photons within a single x-ray pulse is insufficient to form an image with adequate statistics, even at 3rd generation sources. Therefore, time-resolved X-PEEM is accomplished in a pump-probe mode. In such experiments, shown in figure 9, an excitation pulse is synchronized with the incoming x-ray pulses and applied to the sample with a variable delay. Then, by varying the delay and integrating over many repeats of this cycle, the average response of the sample to the excitation pulses can be mapped out in time. When imaging at the highest resolutions, the exposure times are typically on the order of minutes, even at the brightest synchrotron sources. Thus, the resulting images are the average of several tens of millions of pulses.

![Figure 9. Schematic diagram of a pump-probe time-resolved magnetic imaging experiment.](image)

Time varying magnetic fields are generated by current pulses in a lithographic waveguide or stripline, onto which magnetic structures may be deposited directly. A phase relation is maintained by synchronizing the excitation with the x-ray pulses via a timing signal from the storage ring. By varying the phase between the x-ray pulses and the field, the time evolution of the magnetic configuration of the sample can be mapped. The excitation current pulses may be generated electrically, or with pulsed lasers.

For the magnetization dynamics experiments we discuss here, the excitation pulse is usually a fast magnetic field pulse. The first demonstration of pump-probe PEEM magnetic imaging was at the BESSY-II synchrotron in Berlin [101]. In this experiment a microcoil was used to generate field pulses on a NiFe/Cu/Co trilayer, and partial reversal of some of the NiFe domains was demonstrated at a time scale of ~1 ns. While this
experiment demonstrated the possibility of pump-probe PEEM, the use of a microcoil has two significant drawbacks to it. First, the inductance of the coil limits the response time, thus very fast field transitions cannot be coupled to the magnetic structures of interest. In the experiment by Vogel et al. [100], this limited the field rise and fall times to \~10ns. Second, the magnetic field may influence the electron optics, create shifts or distortions in the resulting image, and limiting the spatial resolution.

Typically, it is desired to study the influence of fast field transitions that at least approach the time resolution available \((\sim 100\text{ps})\), and at the spatial resolution available from PEEM \((\sim 100\text{nm})\). To generate such fast field pulses in a way that does not influence the optics, one approach is to place the magnetic structures of interest onto conducting striplines, which are then electrically pulsed. This results in an induced field above the stripline that influences the magnetic structures, but extends only a few microns above them, thus minimizing the effect on the emitted electrons. For ultrafast field transitions requiring high bandwidth, the stripline can be made as part of a coplanar transmission line, which can easily be impedance-matched to the source of the excitation pulses.

Two approaches have been widely used to generate the electrical pulses: the photoconductive switch approach, and direct electrical excitation. In the former, a pulsed laser is used with a DC-biased photoconductive switch integrated into the stripline, thus generating an electrical pulse. This scheme works well where ultrafast \((\text{sub-}100\text{ps})\) unipolar pulses are needed, but is not well suited if an arbitrary waveform is desired. Direct connection to an electronic pulser can in principle be used with any excitation waveform and is easier and less expensive to implement. However, ultrafast pulses may be harder to achieve at the needed repetition rates, and it is very difficult to use in a microscope design where the sample is not electrically grounded.

The synchrotron-based x-ray photoemission electron microscopy (X-PEEM) combines the strength of polarized x-ray absorption enabling both chemical and magnetic sensitivities, electron microscopy enabling high spatial resolution, and the pulsed nature of synchrotron x-ray sources enabling temporal resolution. The chemical sensitivity of PEEM makes this imaging technique uniquely suited to address questions where more than one magnetic element is present. This is particularly useful for investigation of layered nanomagnetic systems of interest for many magnetic device applications, as well as a variety of new, upcoming materials, which have complex chemical and spin structures with the possibility of multiple magnetic elements. The magnetic sensitivity based on the XMLD effect enables the direct imaging of spin configurations in antiferromagnets [102], which can hardly be achieved by any other magnetic imaging method. Time-resolved X-PEEM with high spatial \((\sim 100 \text{nm})\) and temporal \((< 1 \text{ ns})\) resolutions is an excellent tool for studies of spin dynamics in nanomagnetic structures. In the following, we review several recent experiments that demonstrate the significant role of X-PEEM imaging in providing crucial information for fundamental understandings of several problems in nanomagnetism.

4. Recent PEEM Experiments in Nanomagnetism

4.1. Time-resolved studies of vortex dynamics in magnetic disks

Time-resolved X-PEEM is one of the best techniques for directly imaging magnetic vortex dynamics, because time-resolved X-PEEM has the advantages of both the high spatial resolution and the high temporal resolution.

4.1.1. Demonstration of gyrotropic vortex motion in Co squares

In one of the first time-resolved PEEM experiments, Choe et al.[103] used the PEEM-2 microscope at the Advanced Light Source (ALS) to demonstrate the coplanar-waveguide based pump-probe imaging technique, and explored the dynamics of the magnetic vortex state in micron-sized Co structures. Because the PEEM-2 is configured such that the sample must be at the accelerating voltage for the experiment, the authors used a photoconductive switch and a pulsed laser to generate field pulses. This allows relatively fast \((\text{sub-}100\text{ps rise times})\) unipolar pulses.

Co structures were grown on top of the coplanar waveguides using lithographic
techniques, and their sizes and thickness chosen such that the fundamental mode was accessible within the timing constraints of the ALS timing mode used. The structures were demagnetized such that they began with a Landau, or four-domain flux closure state. When subjected to the laser-driven field pulses (300-400ps FWHM), the core of this state is perturbed from the center, and then relaxes towards the center of the square. By varying the delay between the laser pulses and incoming x-ray pulses, the authors mapped the resulting time-dependent domain structure, as shown in figure 10 (a). By analyzing the domain patterns, the location of the vortex core could be derived, and the core trajectories could be plotted, as shown in figure 10 (b).

Choe et al. found that for the magnetic vortices driven resonantly at their excitation frequency, the cores followed a circular (elliptical) path for square (rectangular) shapes, as predicted by the Landau-Lifshitz-Gilbert (LLG) theory. This result was significant because it was the first direct observation of this fundamental gyrotropic motion in the time domain. The authors also confirmed that the sense of the circular core motion (clockwise or anticlockwise about the square center) is dependent only on the polarity of the \( z \)-component of the magnetization of the vortex core, and not on the chirality of the vortex itself (see figure 10 (c)-(e)), as predicted by the LLG theory.

One additional prediction of the LLG theory is the fundamental eigenmode frequency, and its dependence on the geometry of the structure. This was left untested by the ALS experiment, because of the resonant nature of the experiment. Additionally, the use of square structures complicates the application of the analytical LLG theory, because of the introduction of domain walls.

4.1.2. Observation of non-gyrotropic motion in NiFe squares

**Figure 10.** Gyrotropic vortex motion in Co squares of different dimensions. (a) TR-PEEM images of domains in Co squares of Patterns I (1 by 1 \( \mu \)m\(^2\)), II (1.5 by 1 \( \mu \)m\(^2\)) and III (2 by 1 \( \mu \)m\(^2\)), taken at the specified delay times after the field pulse. (b) Trajectories of the vortex core. (courtesy of S-B Choe et al.)[103]

**Figure 11.** Non-gyrotropic vortex motion in 6 \( \mu \)m NiFe squares. (a) time-evolution of the \( y \) component of the magnetization, \( M_y \), averaged over the boxes shown in the right inset PEEM image; (b) same data as in the lower half of (a) but after subtracting the running average together with a fit to the data as described in the ref. [104]; (c) vortex displacement parallel and perpendicular to \( H_p \). The orientation of the exciting field pulse and the polarization \( P \) are sketched in the left inset of (a). (courtesy of J Raabe et al.).

Interestingly, even though it is predicted by a very fundamental and widely applicable LLG theory, the gyrotropic behavior is not universally observed. In a similar experiment at the Swiss
Light Source (SLS) on Ni$_{80}$Fe$_{20}$ alloy (permalloy) squares, Raabe et al. did not observe a circular vortex core trajectory[104], as shown in figure 11. In this experiment, the authors imaged the core motion in 6-µm-square permalloy structure in a flux closure state after a 6mT, ~400ps field pulse generated by a pulsed laser. They found that the core initially moves towards the edge of the square in the direction perpendicular to the field for 1-2 ns. After reaching its maximum excursion of 750 nm, the core relaxes directly back to the center of the square, along essentially the same path. This relaxation takes until the next field pulse, which occurs at $t=16$ ns. The authors also observed precessional motion of the spins within a single domain of the Landau state, and oscillations of domain walls.

The lack of agreement on the presence of gyrotropic motion between the ALS experiment on Co squares and the SLS experiment on permalloy squares is intriguing, since this should be an intrinsic property of a vortex state. Raabe et al. suggest that the difference may be explained by the resonant nature of the ALS experiment as opposed to their experiment, and the higher value of the field pulse amplitude. It is also worth mentioning that the two experiments involved very different materials and different size structures. In addition, both involved squares, which promote the formation of domain walls, for which an analytical solution to the LLG equation does not exist.

4.1.3. Investigation of the free vortex core motion in circular FeNi disks

In circular disks, analytical solutions do exist for the LLG equation, and they make specific predictions for the core trajectories and oscillation frequencies. Thus, circular disks are an ideal platform for exploring the dynamics of the vortex state. Guslienko et al. used time-resolved X-PEEM to investigate the free motion of the vortex core and measure the fundamental eigenfrequencies in the time domain [105]. In this experiment at the Advanced Photon Source (APS), a direct electrical pump pulse was applied, as opposed to a laser-induced field pulse, allowing for an arbitrary time dependent field to be applied. The structures were engineered such that their fundamental frequencies were well above the excitation frequency of 6.53 MHz, determined by the synchrotron bunch structure.

![Figure 12. Free vortex core motion in circular NiFe disks. (a) Time dependence of the ratio of the $y$ displacement of the vortex core to the radius of 5.3 and 4.3 µm diameter dots of thickness 30 nm. (b) Comparison of the XMCD experimental data (symbols) and analytical theory for the dependence of the eigenfrequency of the vortex translational mode on the dot aspect ratio $L/R$ of the cylindrical permalloy dots. [105]](image-url)
oscillation, perpendicular to the direction of the applied field. This result is consistent with those of Raabe et al. [104], but is at odds with the predictions of the LLG equation of motion. The authors suggest, in this case, that the difference may be due to insufficient contrast of the images along the field direction. Since the vortex image inherently has greater contrast in the direction perpendicular to the X-ray beam, a determination of the core position along the beam direction may be subject to greater uncertainties, and that component of the motion may be missed.

4.1.4. Nonlinear effects in vortex dynamics

The observation of non-gyrotropic motion in the free vortex motion of circular permalloy structures rules out the influence of domains walls and resonant driving field as potential explanations for the disagreement with the predictions of theory. Therefore, the possibility remains that the experiments have been conducted outside the range of validity for the theory.

Cheng et al. [107] performed additional experiments at the APS to determine the dependence of the core trajectories on the excitation field amplitude. In this experiment, they found that gyrotropic motion is observed for low excitation fields, while above a critical field of ~2.5 mT, an elliptical trajectory is observed. Thus, they observe the two behaviors noted in the literature within the same experiment, as shown in figure 13. They also observe a transient domain state in the first 500 ps of core motion for high excitation fields, which calls into question the ability to apply linear theory at least to the initial motion.

This critical behavior suggests that a linear regime exists in which the LLG predictions are valid, and above which additional contributions to the dynamics must be considered. Using micromagnetic simulations, the authors reproduced the critical behavior seen in the experiments. The simulations also showed the formation of a cross-tie domain wall corresponding to the transient domain state in the PEEM images. The cross-tie wall includes several vortex-antivortex pairs, which combine with the original vortex as it moves down the wall. This results in many core polarity reversals, effectively randomizing the core polarity. This leads the authors to suggest an alternate explanation for the linear or elliptical trajectories seen above the critical field. They suggest that since the randomization of the core polarity from pulse to pulse in the pump-probe experiment effectively randomizes the sense of the core gyrotropic motion, the component of the circular motion along the beam would be canceled out, and an apparent linear trajectory observed. The same authors later showed that the core polarity and the sense of gyrotropic motion is indeed randomized by large field pulses [108].

Figure 13. Vortex core positions parallel (X) and perpendicular (Y) to the applied field vs time, along with fits (red/solid lines) to damped sinusoids after (a) 1, (b) 2, and (c) 4 mT field pulses. (d) shows the time-dependent applied field profile. The upper inset shows the experimental geometry indicating the relative-field (B) and photonmomentum (k) directions. The lower inset shows the X-deflection/Y-deflection amplitude ratio vs the pulse-field amplitude. (e) Transient domain states observed both in experiment and simulation shortly after the falling edge of a 5 mT excitation pulse (removed at time t=0 ns). The experimental PEEM images (top row) agree well with the micromagnetic simulations (second row). [106, 107]
This explanation depends upon the limited resolution of the imaging experiments. That is, if the imaging technique has sufficient resolution to see both components of the motion, the randomization should be evident as the appearance of two vortex cores at the maximum along the field displacement. The authors above noted a broadening of the core at this point in the motion, but the spatial resolution is insufficient to resolve the two cores. Wiegand et al. [109] performed a similar experiment using a scanning transmission x-ray microscope (STXM) at the Canadian Light Source (CLS). Currently, the STXM technique has a spatial resolution of ~25 nm. Thus, in the CLS experiment, these authors were able to resolve the two cores, and show that for large excitation fields the pump-probe image is a superposition of two gyrotropic trajectories of opposite gyration senses.

4.2. Studies of domain wall dynamics in nanowires

In many previous experimental studies of domain wall dynamics, the domain wall position was determined by indirect means, such as electrical transport measurements [46, 110, 111] and magneto-optical Kerr effect (MOKE) measurements [40, 42]. However, these indirect probing methods cannot provide detailed magnetic configuration of the domain wall (transverse type or vortex type), which is crucial for understanding the fundamental physics underlying the domain wall motion. X-PEEM has been greatly used in imaging magnetic vortex structure because of its sensitivity to in-plane magnetization as well as high spatial resolution. Therefore, it is also suitable for directly imaging the motion of domain wall, especially the vortex type domain wall [112, 113]. More importantly, X-PEEM combining with the coplanar waveguide technique can be used in a pump-probe arrangement for time-resolved study of domain wall dynamics [114]. X-PEEM imaging at various temperatures can also study temperature dependence of the domain wall dynamics [113].

Thomas et al. [112] used X-PEEM to explore the injection and pinning of magnetic domain walls in a 250-nm-wide, 20-nm-thick Ni_{80}Fe_{20} nanowire with a micron-sized elliptical nucleation pad [115, 116] at one end and the other end tapered to a sharp tip [117, 118]. The nanowire itself is straight apart from one or two triangularly shaped notches formed at the edge of the wire to trap domain walls [118, 119]. Direct PEEM imaging revealed that a vortex-like structure is readily nucleated in the nucleation pad at low magnetic fields (<15 Oe), whereas injection of a domain wall into the nanowire requires significantly larger fields (~60 Oe). The authors also found domain walls can be pinned in the nanowire at the notches and the chiralities of the vortex-like domain walls vary in successive experiments. These results show the subtle complexities of the domain wall nucleation, propagation, and pinning processes.

Laufenberg et al. [113] performed X-PEEM imaging to determine the spin structure of head-to-head domain walls in Ni_{80}Fe_{20} rings of various dimensions and further extracted the corresponding room temperature phase diagram which exhibits two phase boundaries between the wall types. In temperature-dependent X-PEEM imaging as shown in figure 14, the authors observed a thermally activated switching from transverse walls to vortex walls at elevated temperatures at a transition temperature between 260 °C and 310 °C. This provides direct experimental evidence for the fact that transverse and vortex walls are separated by an energy barrier which can be overcome thermally.

Figure 14. PEEM images of a 7 nm thick and 730 nm wide ring imaged during a heating cycle at temperatures of (a), (d) T=20 °C (before and after heating, respectively), (b) T=260 °C, and (c) T=310 °C. The two transverse walls (a) are not visibly influenced by heating (b) up to the transition temperature (c), at which a thermally activated transition to a vortex type occurs in both walls. (d) The vortex walls are retained after cooling down. The gray scale shows the magnetization direction. [113]
X-PEEM has also been used to investigate current-driven domain wall motion. Kläui et al. [120] reported the effect of electrical currents on the propagation and spin structure of vortex walls in NiFe wires. They found that in the 1 μm wide and 28 nm thick NiFe wires, the movement of unperturbed vortex walls started at around 8.3x10^{11} A/m² and the average wall velocity increases with increasing current density, and the typical velocity is below 1.0 m/s. In addition to propagation of unperturbed vortex walls, they also observed a number of different wall transformations. Vortices were found to be nucleated (as shown in figure 15) and annihilated due to the spin torque effect, in agreement with theoretical predictions [121]. The velocity was found to be directly correlated with these transformations and decrease with increasing number of vortices. Uhlir et al. [122] demonstrated very high velocities for current-induced domain wall propagation in NiFe/Cu/Co trilayered nanowires with maximum velocities exceeding 600 m/s. These velocities are 4 to 5 times larger than the maximum value reported for other in-plane anisotropy systems [123].

Figure 15. (a) PEEM image of a vortex wall after remagnetization (top) and after injection of a pulse with j=8.3x10^{11} A/m² (bottom). An extra vortex has been nucleated downstream. (b) A high resolution image of the double vortex wall and a simulation with arrows (c) visualizing the spin structure. (d) Image of an extended vortex wall with a cross-tie structure at the center and arrows indicating the magnetization direction. [120]

Recently Uhlir et al. [114] have used time-resolved X-PEEM to investigate the magnetization dynamics induced by nanosecond current pulses in NiFe/Cu/Co nanowires. They found that The Oersted magnetic field present during the current pulses induces a large tilt of the NiFe magnetization, transverse to the wires, as shown in figure 16. They also observed spin-wave-like oscillations of the NiFe magnetization and attributed these oscillations to precessional motion about the effective field. Their results clearly show that the effects of the Oersted field should be taken into consideration when interpreting current-induced domain wall motion in multilayered nanowires.

Figure 16. Time-resolved PEEM images of the NiFe layer of a 400 nm wide nanowire at time delays of (a) 0 ns, (b) 0.35 ns, (c) 0.45 ns, (d) 1.9 ns, (e) 2.2 ns, (f) 2.3 ns, (g) 2.4 ns, (h) 3.3 and (i) 3.6 ns with respect to the beginning of the positive part of the bipolar current pulse. These delays are indicated on the bipolar pulse plotted in (j), together with the magnetization tilt angle. [114]

4.3. Studies of exchange bias in ferromagnet-antiferromagnet interfaces

Element-specific X-PEEM imaging techniques based either the XMCD effect or XMLD effect allow the observation of the microscopic spin structure on both sides of an ferromagnet-antiferromagnet interface, providing a extremely powerful tool for exploring a hotly debated question: the origin of the exchange bias in the ferromagnet-antiferromagnet system. Here
we review the first direct imaging investigation of interfacial magnetic coupling in an exchange bias system X-PEEM, followed by a discussion of the importance of considering the anisotropy of XMLD signals and the review of X-PEEM studies of interfacial uncompensated spins of the antiferromagnet.

4.3.1. The first X-PEEM observation of interfacial magnetic coupling in exchange biased layers

In one of the first experiments to exploit the element selectivity of PEEM in a layered system, Nolting et al [124, 125] explored the interfacial magnetism in exchange-biased bilayers. In this experiment, the authors used x-ray magnetic dichroism in PEEM to image the antiferromagnetic and ferromagnetic domain structures of the Co/LaFeO3 bilayer. The authors compared the linear dichroic images observed in the antiferromagnetic LaFeO3 layer at the Fe L3 resonance with the corresponding circular dichroic images at the Co L3 resonance, as shown in figure 17.

The authors discovered a clear correlation of the ferromagnetic Co domains and the antiferromagnetic LaFeO3 domains, indicating a strong exchange coupling at the interface between the two layers. They demonstrated that the exchange bias occurred on a domain-by-domain basis, confirming its microscopic origins. This domain-by-domain exchange coupling is critical to the formation of the exchange bias effect. They also attempted to image the uncompensated Fe spins directly with XMCD microscopy, but did not succeed. They thought it was probably because of the small concentration or magnitude of the uncompensated spins.

4.3.2. Importance of experimental geometry in interpreting the XMLD signal

In a follow up experiment to Nolting, et al. [124], Czekaj et al. [126] examined the geometrical dependence of the linear dichroism signal in PEEM images of LaFeO3. In this experiment, they continuously varied the angle between the incoming photon polarization and the antiferromagnetic domain magnetization directions. In doing so, they were able to map out the full angle dependence of the XMLD signal, and they found that the magnetization directions were aligned ~20° away from the crystalline axes and the film plane. More importantly, they showed that the XMLD signal depends strongly on the magnetocrystalline anisotropy, and that it can change sign, depending on the experiment geometry.

Taking into account the anisotropy of XMLD signal is very important for a correct interpretation of antiferromagnetic domain images [127, 128]. For example, Arenholz et al [129] and Ohldag et al [130] recently reported that the magnetic spins near the surface of a NiO(001) single crystal are perpendicular to those in the Co thin film deposited on top of this NiO layer. These results are based on a comparison between the XMCD image of the Co layer and the XMLD image of the NiO layer, which carefully took into consideration of the XMLD anisotropy on the intensity ratio of the two peaks at the Ni L2 edge. This perpendicular alignment of the ferromagnetic and antiferromagnetic spin in the Co/NiO system is contradictory to an earlier report of a perfectly parallel alignment of the ferromagnetic and antiferromagnetic spins in the same system [131], which did not take the anisotropic XMLD into account. This shows that the relative orientations of the crystalline axes, film plane, and photon polarization must be well understood for the

Figure 17. PEEM images and local spectra from the antiferromagnetic and ferromagnetic layers for 1.2-nm Co on LaFeO3/SrTiO3(001). (a) Fe L-edge XMLD image; (b) Co L-edge XMCD image. The spectra shown underneath were recorded in the indicated areas and illustrate the origin of the intensity contrast in the PEEM images. [124]
interpretation of antiferromagnetic domain images, and for understanding exchange bias effects.

4.3.3. Studies of uncompensated spins in exchange-biased and coupled bilayers

Extensive research results suggest that the exchange bias effect originates in uncompensated and the pinned magnetic moments of the antiferromagnet layer [15, 132, 133]. The high sensitivity of XMCD enhanced by dividing images taken with left and right circularly polarized light allows probing such uncompensated spins even if their effective thickness is below a monolayer.

Eimüller et al. reported the direct XMCD imaging of uncompensated Mn spins at the CoFeB/MnIr/NiFe system taken with PEEM at the (a) Co and (b) Mn L3 edges. Arrows in some domains of (a) represent their magnetization direction as deduced from the XMCD contrast [134].

Eimüller et al reported the direct XMCD imaging of uncompensated Mn spins and the at the CoFeB/MnIr interface with an antiferromagnetic coupling between the Mn and the Co magnetic moments, as shown in figure 18 [134]. A square region of the CoFeB/MnIr film where material was removed by focused ion beam (FIB) etching, except for quadratic dots with edges of 1, 1.5, and 2 μm length were imaged by X-PEEM at Co and Mn L3 edges. The magnetization of most Co domains points along the exchange bias direction with a spread of orientations relative to this direction of less than 45°. The same domain structure observed for Co was also observed at the Fe L3 edge (not shown), thus proving parallel coupling of Co and Fe spins. For the disordered γ-phase fcc alloy Mn77Ir23 a vanishing XMCD is expected since its magnetic Mn moments are compensated in the (111) film plane, forming a noncollinear, so-called 3Q spin density wave (SDW) structure [135]. However, at the CoFeB/IrMn interface, the same domain structure found in CoFeB was also observed when the photon energy was tuned to the Mn L3 edge, but with a reversed contrast, indicating an antiparallel alignment between the uncompensated Mn and the ferromagnetic CoFeB spins.

Baruth et al. [136] also observed interfacial uncompensated spins of an antiferromagnet in a trilayer with a single NiO layer sandwiched by two Co/Pt multilayers with perpendicular magnetic anisotropy [137, 138]. Interestingly, an oscillatory coupling was observed in this system by Liu et al. through the insulating antiferromagnetic NiO spacer [139, 140]. Magnetic coupling in layered structures has been studied intensely ever since the discovery of antiferromagnetic oscillatory coupling through nonmagnetic layers, and the concurrent GMR effect [55]. Most of work has been on all-metallic systems, where an RKKY mechanism was invoked to explain the oscillatory interlayer thickness dependence [141]. For insulating spacers, an oscillatory dependence was not predicted [142, 143].

To investigate the origin of the oscillatory coupling in this system [Co/Pt multilayer]/ NiO [Co/Pt multilayer] system, Baruth et al. using soft x-ray spectroscopy and X-PEEM imaging at the Ni and Co L edges. By comparing the Co and Ni L edge XMCD signals, Baruth et al found a non-zero Ni moment that was correlated with the sign and strength of the observed interlayer coupling. For antiferromagnetic Ni, no XMCD is expected since the net spin is zero. Therefore the observed signal suggests that uncompensated spins at the Co/NiO interfaces cant in the perpendicular direction as a result of the Co-Ni exchange coupling. This canting may be transmitted through the NiO layer to the opposite Co layer, thus creating an indirect coupling. The authors showed this net Ni moment first in XMCD spectroscopy, however a crucial test of whether it truly originates from interfacial spins was performed by X-PEEM imaging at the Co and Ni L3 edges. The PEEM images revealed that the Co and Ni spins follow each other in a domain-by-domain fashion, proving the microscopic nature of the coupling.

Miguel et al. [144] recently examined both the ferromagnetic and antiferromagnetic domain structure in an Fe/CoO exchange biased bilayer system. Using the XMCD and XMDL effects at the Fe and Co L3 edges, they were able to image the Fe ferromagnetic domains, and Co
uncompensated spins, and the Co antiferromagnetic domains separately, as shown in figure 19.

Miguel et al. found that the Co uncompensated spins are coupled ferromagnetically to the Fe spin by the strong exchange coupling. Further, the Fe and Co ferromagnetic spins are collinear with the Co antiferromagnetic spins. The authors showed also that the antiferromagnetic domain structure is strongly influenced by the ferromagnetic ordering of the Fe layer. For no Fe coverage, the Co XMLD contrast is barely recognizable, suggesting that the domains are smaller than the spatial resolution of the microscope. For 0-2 monolayers of Fe coverage, below the critical thickness for FM ordering, there is also very little XMLD contrast. Above this critical thickness, the Fe layer orders very clearly, and antiferromagnetic domains appear that are correlated with the FM domains. This correlation provides important clues to the mechanism that underlies the exchange coupling.

4.4. Studies of spin reorientation transitions in coupled magnetic multilayers

The full-field and element specific natures of the PEEM technique in conjunction with a wedge growth technique are uniquely suitable for investigating the spin reorientation transition (SRT) in ultrathin films or magnetic multilayers [70, 145, 146, 147, 148, 149, 150]. This was first demonstrated by Kuch et al [70] in a study of SRT in fcc wedged Co/wedge Ni/Cu(001) epitaxial ultrathin films as a function of Co and Ni film thickness. The Co thickness was varied from 1.4 to 2.6 atomic monolayers (ML) and the Ni thickness from 11 to 14 ML. Both Co and Ni layers were prepared as crossed wedges with slopes rotated by 90° with respect to each other. The authors acquired a series of PEEM images at different photon energies which cover the Ni 2p3/2 and 2p1/2 photon absorption regions. Therefore in addition to the qualitative information about magnetic domains obtained at one fixed peak photon energy, full quantitative information can be extracted from XMCD analysis of such a spectral series of images with the same spatial resolution.

Figure 19. XMCD–PEEM magnetic domain images of 5 monolayers Fe at room temperature (a) and at low temperature (b). Magnetic domain image originating from uncompensated Co moments at the interface as seen by Co L3 XMCD–PEEM (c) and antiferromagnetic domains of the 8 monolayers CoO layer by Co L3 XMLD–PEEM (d), both at low temperature. The red lines are guides to the eye to follow the overlap of Fe and CoO domain walls. [144]

Kuch et al showed that the spin reorientation occurs as a function of both Co and Ni thicknesses. A switching of the magnetic easy axis from [001] out-of-plane to <110> in-plane directions when increasing the Co thickness or decreasing the Ni
thickness. Pixel-by-pixel sum-rule analysis of the series of PEEM images results in images displaying the Ni spin and orbital magnetic moments, projected onto the propagation direction of the illuminating x-rays, as shown in figure 20. A constant effective Ni spin moment similar to the bulk magnetic moment was observed. However, the Ni orbital moment is distinctly higher for out-of-plane magnetization than for in-plane magnetization. The domain density of the perpendicular magnetization increases towards the spin reorientation transition line. The authors explained the formation of perpendicularly magnetized domains with decreasing size upon approaching the spin reorientation transition by magnetostatic stray field energy minimization for decreasing domain wall energy. This experiment help to elucidate the detailed mechanism of the spin reorientation transition in the Co/Ni/Cu (001) system.

Figure 21. (a) SRT position in the Fe-Ni thickness plane. Magnetic bubble domain images of (b) \(d_{Fe} = 2.9\) ML, \(d_{Ni} = 5.5\) ML; (c) \(d_{Fe} = 3.1\) ML, \(d_{Ni} = 8.0\) ML; (d) \(d_{Fe} = 3.6\) ML, \(d_{Ni} = 10.8\) ML; (e) \(d_{Fe} = 4.1\) ML, \(d_{Ni} = 13.6\) ML. [149]

Choi et al. [149] used PEEM imaging in conjunction with a wedge growth technique to examine thickness-dependent changes in magnetic domain structure as a thin film system is driven through a spin reorientation transition. In their experiment, the influence of the anisotropy on the formation of bubble domain phases in thin films with perpendicular magnetic anisotropy was examined in a crossed wedge Fe/Ni bilayer structure deposited on a Cu(001) surface. By varying the Fe and Ni thicknesses in orthogonal directions, they created a 2D continuous library of every thickness combination. This then allowed them to map out the response of the system to the thickness-driven changes in anisotropy by imaging the wedge structure with X-PEEM, as shown in figure 21. They found that a new bubble domain phase forms near the thickness at which the magnetic anisotropy switches from out-of-plane to in-plane. This domain phase turns out to be a metastable phase, appearing only with a small out-of-plane field, and disappearing if the sample is heated above 370 K.

4.5. Study of magnetization switching by electric field in ferromagnet-multiferroic structures

Element-specific and layer-resolved X-PEEM technique has recently been applied to study ferromagnet-multiferroic bilayers. Chu et al [72] investigate a bilayer consisting of a ferromagnetic Co\(_{0.9}\)Fe\(_{0.1}\) in intimate contact with a multiferroic BiFeO\(_3\) layer. The magnetic state of the CoFe layer was imaged by X-PEEM based on the XMCD effect at the Co L-edge. The authors determined that the CoFe forms domains that are aligned 90\(^\circ\) with respect to each other and that the local magnetic domain structure exactly follows the ferroelectric domain structure of BiFO\(_3\) imaged by piezoresponse force microscopy (PFM), as shown in figures 22 (a) and (b). These images provide the first direct microscopic evidence of exchange coupling between a ferromagnet and a multiferroic at this length scale (~200 nm). The authors attributed the observed one-to-one mapping of the ferroelectric and ferromagnetic domains to the collinear coupling between the magnetization in the ferromagnet and the projection of the antiferromagnetic order in the multiferroic.

The authors further used X-PEEM to directly image the change in magnetic state of the CoFe layer in response to the electric switch of the multiferroic BiFeO\(_3\) layer, as shown in figure 22 (c)-(e). Their observations indicate that the average magnetization direction in the ferromagnet rotates by 90\(^\circ\) on the application of an electric field to the underlying BiFeO\(_3\) layer. On electrical switching the BiFeO\(_3\) layer once again, the average magnetization direction changes back to the original state. Their results demonstrated the possibility to locally control ferromagnetism with an electric field.
4.6. Magnetization reversal in complex nanostructures

4.6.1. Studies of ferromagnet - multiferroic nanocomposit films

In addition to heterostructures consisting of ferromagnetic and multiferroic layers, ferromagnets and multiferroics can also be integrated in the format of nanocomposit films containing magnetic nanopillars embedded in a multiferroic film. X-PEEM has demonstrated its unique strength of element specific magnetic imaging in this type of complicated, multi-phased nanomaterials. Zhao et al [151] studied the magnetic structure as well as its response to an external electric field in ferrimagnetic CoFe$_2$O$_4$ nanopillars embedded in an epitaxial ferroelectric BiFeO$_3$ film using XMCD-based X-PEEM imaging. The film was first magnetized in an out-of-plane magnetic field, resulting in a predominant downward magnetization. Then, an electric voltage of $-12$ V was applied in order to switch the ferroelectric polarization in the BiFeO$_3$ matrix by scanning the PFM tip over a selected area. Subsequent PEEM imaging was performed in an area containing both poled and unpoled regions. Magnetic switching was observed in both Co and Fe magnetic sublattices of the ferromagnetic CoFe$_2$O$_4$ nanopillars after application of an electric field. The XMCD-based PEEM image showed only black dots, indicating a uniform downward magnetization distribution, in the unpoled region (outlined by a box with red solid lines in figure 23 (a)), while both black and white dots can be seen in the poled area (outlined by a box with green dashed lines in figure 23 (a)), indicating switching of some of the nanopillars. From XMCD spectroscopy experiments, about 50% of the CoFe$_2$O$_4$ nanopillars were measured to switch their magnetization with the electric field, implying an elastic-mediated electric-field-induced magnetic anisotropy change.

Figure 23. Co-edge PEEM images at the interface between poled and unpoled regions. (a) Magnetic information. (b) Chemical information. The scale bars are 1 µm. [151]

To ascertain that the dichroism contrast seen in the PEEM image originates from the CoFe$_2$O$_4$ nanopillars, a Co element-contrast PEEM image was obtained by taking the sum of two images acquired on the left shoulder of the Co L$_3$ peak.
with right and left circular polarizations, as shown in figure 23 (b). By comparing figures 23 (a) and (b), the authors showed that the areas showing magnetic contrast correspond to areas containing Co. The combination of the magnetic information and chemical information, obtained by X-PEEM, of the unpoled and poled areas in the nanocomposite CoFe₂O₄-BiFeO₃ film convincingly demonstrated the magnetization switching of CoFe₂O₄ nanopillars by the electric field.

4.6.2. Studies of multilayered nanodisks

The ability of PEEM to provide element-specific magnetic images has proved very useful for understanding the behavior of ferromagnetic spin valve structures. There is interest in using metallic trilayers, such as NiFe/Cu/Co, patterned into ring-shaped nanostructures, for magnetic memory elements. The ring-shaped geometry promotes the formation of vortex states, which are useful for minimizing the stray fields from the memory element, thus allowing greater density. However, there is evidence that the layered geometry may influence the formation of the vortex states via the dipolar magnetic interactions between the layers,[152] and micromagnetic simulations suggest a complicated phase diagram.[153]

Rose et. al. [78] used PEEM to image the magnetic domain states of the permalloy layer in NiFe/Cu/Co trilayer rings, and compared the observed images with layer-specific hysteresis loops obtain by resonant x-ray magnetic reflectivity. They found that although the hysteresis loops showed zero remanent moment, suggesting the formation of vortex states, the PEEM images of the top NiFe layer showed a multidomain structure, as shown in figure 24.

While the detailed domain structure displayed some randomness, a four-domain pattern appeared with greater frequency than would accounted for by chance. Comparison of these multidomain patterns with micromagnetic simulations suggests that they are actually antivortex states. This experiment therefore shows the importance of imaging the real-space domain structure, even in cases where layer-specific hysteresis loops on an ensemble of nanostructures are available.

4.6.3. Studies of an artificial spin ice system

The kagome spin ice arrangement has been of particular interest because it is highly frustrated and the three interactions at a vertex are equivalent. The full-field nature of the PEEM technique makes it uniquely suitable for investigating a large array of magnetic nanostructures, such as an artificial spin ice system [86, 154]. Mengotti et al. [86] used X-PEEM to examine the magnetic configurations in three basic building blocks of an artificial kagome spin ice consisting of one, two, and three rings. The XMCD-based PEEM images exhibit a high magnetic contrast with four distinct gray levels associated with the kagome structure, allowing one to determine unambiguously the direction of the moments in all islands with a single image, as shown in figure 25. Following a demagnetization process involving rotation of the sample in a magnetic field, the authors determined the frequency of states as a function of the dipolar coupling strength between the islands, modified by varying the lattice parameter. They found the important result that, as the number of rings increases, there was a dramatic reduction in the ability to achieve the low-energy states via demagnetization.

Figure 24. PEEM image of the NiFe layer of the multilayer rings exhibits diverse multi-domain structures at remanence. This suggests that imperfections in the rings strongly control the magnetic reversal. [78]
They also directly examined the magnetization reversal in an applied field in situ in the PEEM. Following saturation of the magnetization, they recorded XMCD snapshots of the switching process on applying a reverse field, increasing its value from zero in steps of approximately 6 Oe, and recording images at remanence after reducing the field between each step back to zero. Their in situ magnetization reversal imaging results also showed that as the number of rings increases there is a drastic decrease in the ability to achieve the low-energy states. This PEEM imaging study of magnetic configurations in three basic building blocks of an artificial kagome spin ice provides crucial information for understanding magnetic frustration in an artificial spin ice system.

5. Future Prospects

In the preceding, we have outlined the recent progress in static and time-resolved magnetic imaging using PEEM over the last 10–12 years. All of this work has been based on the standard electrostatic PEEM design in conjunction with a helical undulator at a 2nd or 3rd generation synchrotron source. However, new sources and electron optical designs are currently coming online or are under development that promise to provide orders of magnitude better performance in sensitivity and spatial and temporal resolution. Principle among these developments are the use of aberration-corrected electron optics, which would allow spatial resolution down to 2–5 nm, and bunch slicing or compressing schemes to improve the time resolution to sub 1 ps. In the longer term, 4th generation sources (x-ray free electron lasers) are now beginning to become available, and may produce opportunities for entirely new types of PEEM experiments in the future.

5.1. Aberration-corrected optics for improved spatial resolution

In the most typical PEEM optical setup, the spatial resolution is limited by aberrations in the electron optics. The two most significant contributions to these are chromatic aberration due to the finite energy spread of photoelectron emitted from the surface, and spherical aberration. For excitation with soft x-rays, chromatic aberrations effectively limit the spatial resolution to ~100 nm. In the absence of chromatic aberrations, spherical aberrations would limit the resolution to ~20 nm. Indeed, this spatial resolution has been achieved in PEEM systems with UV excitation, in which the electron energy spread is limited by the lower energy of the incoming photons[155]. In the absence of any aberrations in the optics, the spatial resolution is limited by the effective area from which electrons may be emitted. Since most of the electrons emitted are secondary electrons, this area will be

![Figure 25](image)

**Figure 25.** Experimental frequency of low-energy states (band 1) for strongly coupled islands with increasing number of rings. (a) Geometries for demagnetization with in-plane and out-of-plane rotation axes and for in situ switching in the PEEM, (b) XMCD images of nine of the one-, two-, and three-ring configurations following demagnetization about an in-plane rotation axis, and (c) the frequency of observed states plotted against the number of rings. [86]
defined by the mean free path in the material being imaged. In most cases, the effective limit for the resolution is 2-5 nm.

There are currently two efforts to build improved resolution PEEM instruments at x-ray facilities through various levels of aberration correction. The first one, at BESSY-II, is called SMART[156], and the second is the PEEM-3 instrument at ALS [157]. To reduce chromatic aberrations, an energy filter is typically used to restrict the energy distribution of the electron beam. To correct for spherical aberrations, the beam is deflected into an electron mirror which introduces equal and opposite aberrations. If Fig. xx, we show a schematic diagram of this approach for the PEEM-3 instrument at ALS.

5.2. Short pulse sources at storage rings for improved time resolution

In standard storage ring-based pump probe experiments, the ultimately achievable time resolution is governed primarily by the length of the electron bunches in the ring. Other effects may degrade the resolution from there, such as timing jitter and reduced bunch purity, but the bunch length clearly places a lower limit on the resolution. While it is physically possible to generate electron bunches in the 100 fs regime, the use of storage rings limits the steady state bunch length through Coulomb broadening. Therefore, the bunch length is dependent on the storage ring design, the bunch pattern used, and the charge per bunch. At most 3rd generation sources, the available bunch lengths range from ~50 – 100 ps (FWHM). For much field-driven magnetization dynamics research, 100 ps resolution is sufficient. However, faster phenomena occur, in particular optically-driven effects, at time scales from 100 fs to 10 ps. One such example is thermal demagnetization, which can take place in sub-ps time scales, and is of interest for thermally-assisted magnetic recording technology [158].

Direct coupling of the magnetization to a laser field can also produce magnetization dynamics in the sub-ps to 10’s of ps scales [159]. There are several potential methods to improve the pulse length available at x-ray sources. These involve either modifying the electron bunches to allow a shorter “slice” of the x-ray pulse to be picked out, or using next generation sources.

Of the former, the most widely used approach is the femtosecond laser slicer. In the laser slicing technique, ultrafast x-ray pulses are created using the interaction of a pulsed laser and the electron bunches in the storage ring [160]. Laser pulses are launched into the storage ring tube, where their strong electric field modulates the energy of a small slice of the electron bunch. This creates a dispersion in the emitted photons from that bunch, such that the resulting short x-ray pulse may be separated from the rest of the pulse using a slit. The laser slicing method has the advantage that it is capable of generating very fast x-ray pulses (~100 fs). However, the cost in available flux is very high. First, by slicing 100 fs out of a 100ps bunch, a factor of 10^3 is thrown away. Also, the pulsing laser usually can only be operated at ~1kHz, such that all other x-ray bunches that are not subject to a laser pulse must be gated out. As storage ring repeat rates are usually above 1MHz, this results in a loss of an additional factor of 10^7. Therefore, the flux obtained from femtosecond slicing beamlines is usually only about 10^4 ph/s/0.1% bandwidth, compared with 10^{12} for a beamline designed for XMCD spectroscopy and PEEM that can count every bunch. The capability to do time resolved imaging with such low flux numbers is extremely limited, but is possible in principle. It was recently demonstrated by Boeglin et al. that high quality XMCD spectroscopy, sufficient for quantitative analysis of the spin and orbital moment dynamics, is possible with the laser slicer at BESSY-II [161]. With sufficient stability and access to beamtime, a PEEM experiment that involves fixed-energy imaging could be possible, although to date none have been reported.
Besides laser slicing, other approaches to manipulating the electron bunches within the storage can produce high flux and significant improvements in the time resolution to the 1 – 10 ps range. At the BESSY-II light source, a special bunch compression technique known as low-\(\alpha\) mode provides <4 ps soft x-ray photon pulses, with a bunch spacing of \(\approx 2\) ns, and about 10x the charge per bunch as in laser slicing [162]. Since this bunch compression technique can be used with every bunch, it yields significant improvements in average flux over laser slicing. Schönhense et al., used this special mode in a field-driven pump-probe PEEM experiment on micro-scale NiFe square and rectangular structures[163], obtaining a time resolution in imaging of 15 ps [164]. These experiments revealed several transient features of the magnetization process, including blocked domains and transient domain walls and vortices. As in the vortex studies of Guslienko et al. [105] and Cheng et al. [107, 108], these states persist for <1 ns, and do not exist in quasistatic magnetization processes.

The low-\(\alpha\) mode requires very low charge per bunch, therefore in order to obtain the high average flux needed for the PEEM experiments and other user experiments, the repeat rate is kept high, and the bunch spacing short. This limits the total time available to follow magnetization processes before the next pump pulse comes. Thus the low-\(\alpha\) mode offers improved time resolution, but has a limited dynamic time range. Another proposed scheme for bunch manipulation may offer very high time resolution, high average flux, and repeat rates that approach normal synchrotron operating modes. In this idea, a special rf cavity is used to modify the momentum profile of the bunch prior to their entry into an undulator, such that the emitted photons from the upstream and downstream ends of the bunch diverge [165]. A horizontal slit is then used to take a time slice out of each bunch, and a second cavity downstream of the undulator restores the particle beam characteristics, as shown in figure 27. This technique can, in principle, be used at high repeat rates with cryogenic cavities, while the storage ring may be operated in a standard configuration. It therefore offers potentially high average flux with no impact to other users at the facility. This approach has not been implemented at any facility as of this writing, but is proposed for the Advanced Photon Source in an upgrade plan intended for completion in 2017. Estimates for the proposed APS short pulse x-ray source (SPX) suggest that pulse lengths of \(~1\) ps can be obtained for hard x-rays, and \(~4\) ps for soft x-rays, with \(~10^8\) ph/s/0.1% bandwidth [166]. This type of source would be very useful for studying sub-10 ps magnetization phenomena, as occur in semiconductor spin dynamics and laser-driven pump-probe experiments.

**Figure 27.** Bunch-deflection scheme of Zholents [165] for generation of short x-ray pulses under consideration at the Advanced Photon Source.

### 5.3. Fourth generation x-ray sources

For improvements in time resolution beyond 1 ps, a move beyond standard storage-ring based x-ray sources is necessary. The most widely implemented approach to this so far is the free-electron laser (FEL), which produces ultrafast pulses with very high peak flux, although usually with low repeat rates. Two such facilities currently exist, the Free Electron Laser in Hamburg (FLASH) soft x-ray FEL at Hasylab, Hamburg, Germany[167], and the Linac Coherent Light Source (LCLS) hard x-ray source at the Stanford Linear Collider (SLAC) in California, USA[168]. Both of these facilities provide \(10^{12} – 10^{13}\) photons/pulse (equivalent to \(~10\) s, or \(10^7\) pulses, of operation at a typical storage ring) with pulse durations as low as \(10-50\) fs. The photon energies available are from the UV to 180 eV at FLASH, and 800 eV to 8.3 keV at LCLS as of this writing. In future, LCLS may be able to provide energies down to 500 eV.

The available energy range of LCLS covers, or will cover, most of the important edges in elements important for magnetism. Therefore, although the available polarization is currently limited to linear, it is worthwhile to briefly consider how a hypothetical circularly polarized source at this facility would operate and contribute
to magnetism research. The average flux at LCLS at 120 Hz is \( \approx 10^{15} \) photons/s/1% bandwidth, an improvement of only 10x over 3rd generation storage rings. However, the pulse duration and the peak flux allow this source to access new regimes and new physics that are unattainable at storage ring sources.

The most obvious advancement comes from the ultrashort pulse length, as low as 10 fs (FWHM). Absorption spectroscopies at this time scale would enable, for the first time, magnetization dynamics of sub-ps processes. This would then allow probes of how atomic and molecular transitions couple to the magnetization. The coupling of ultrafast laser fields has been shown to induce dynamics in the sub-ps time scale in certain oxides\(^\text{[55]}\) which are not fully understood.

The ability to image at less than 100 fs would undoubtedly produce outstanding results using the same pump probe approaches as needed at synchrotrons, and lead to new magnetization physics at new time scales. However, the most significant advancement from using such a source will likely be the ability to go beyond stroboscopic techniques. The flux per pulse of the LCLS is equivalent to \( \sim 10 - 50 \) s of beamtime at a state-of-the-art, 3rd generation soft x-ray synchrotron beamline. A typical PEEM exposure time may be 10 s to 5 min, therefore the FEL is the first source available that is able to form a useable PEEM image from a single pulse. This removes a major restriction of pump-probe imaging: its insensitivity to any stochastic domain behavior.

Because stroboscopic imaging averages several tens of millions of pulses, any behavior that does not happen the same way with extremely high repeatability will be missed. For most device applications, non-reproducible behavior is of course undesired, so pump-probe techniques should work well. However, it is important to understand the non-repeatable behaviors, as they may appear unexpectedly in certain geometries, or they may be an unavoidable consequence of the geometry or the driving excitation. For example, in the motion of a domain wall along a narrow nanowire above the Walker limit, the complex vortex-oscillation driven motion may have a strong stochastic component. In general, the motion of a magnetic vortex above its critical velocity is characterized by a high degree of core distortion, the formation of vortex-antivortex pairs, and random core polarity reversals.

While a PEEM experiment at an FEL facility would offer the ability to probe these stochastic dynamical phenomena in a new way, it would be subject to the same constraints that other FEL experiments have come up against. The first is that the peak power in each pulse is extremely high, and may result in extensive damage to the sample, especially for insulating samples. Therefore, rather than an ensemble of experiments on a single sample, a typical FEL experiment may involve a series of single shot experiments on an ensemble of samples. This brings in new considerations for creating reproducibility across samples, to allow for systematic comparisons to be made.

### 6. Summary

In the field of nanomagnetism, the important length scales can range from the exchange length (of order 10 nm) to the characteristic domain size (several microns or more). Dynamical behavior occurs at very fast timescales, ranging from sub-ps to microseconds. Therefore, progress in this area depends strongly on characterization techniques, especially direct magnetic imaging, that offer high sensitivity, high spatial resolution, and high time resolution. Using photoemission electron microscopy with excitation by circularly polarized soft x-ray sources, images with 100 nm spatial resolution and very high magnetic sensitivity can be obtained by tuning to an appropriate absorption resonance. At the same time, the timing structure of synchrotron sources is often ideal for magnetization dynamics, enabling experiments in a pump-probe mode. Consequently, PEEM experiments have made many important contributions to the field of nanomagnetism over the last 20 years.

Earlier PEEM magnetic imaging experiments concentrated on understanding the relative magnetic order in several layered systems, in which multiple chemically distinct magnetic components are present. Here, the element specificity inherent with soft x-ray excitation provides definitive information about the spin configuration, and therefore robust models of the interfacial spin coupling can be developed and tested. In addition to sensitivity to ferromagnetic layers, X-PEEM provides sensitivity to
antiferromagnetic order in oxides and other localized electron systems via directional coupling of incident photons to multiplet states. Such experiments have been very instructive in examining the domain relation between antiferromagnetic and ferromagnetic components in exchange biased systems. These may also be correlated with uncompensated interfacial spins for a more complete understanding of the interfacial spin structure. The orbital sensitivity obtained with X-PEEM also provides directional information, making it extremely useful for exploring magnetic anisotropies.

Another very useful aspect of PEEM with synchrotron excitation is the inherent time resolution that may be obtained from the pulsed nature of the x-ray source. At most 3rd generation storage rings, the timing structure of the electron bunches results in x-ray pulse durations of 50 – 100 ps, with ~2 – 150 ns in between bunches. Given that important magnetization dynamics processes occur in frequency ranges from MHz to THz, this timing structure is uniquely suited for time resolved imaging of magnetic nanostructures, using a pump-probe approach. Time-resolved X-PEEM has been applied to the dynamics of magnetic vortices in small (1 μm-sized and larger) lithographic structures. These experiments have yielded important understanding of the vortex fundamental gyrotropic mode frequency, its relation to shape, and the influence of non-linear effects on the vortex core trajectories. Time resolved PEEM has also been applied to the questions of domain wall motion in nanostructures and nanowires, and magnetization reversal.

To date, most PEEM experiments on magnetic structures have involved objects of ~500nm size and larger, owing to the typical resolution of 100nm. In future, many magnetic device applications will involve smaller dimensions and faster dynamics that go beyond the present limits of pump-probe PEEM. In addition, the use of newer, more anisotropic materials in such devices may pose a challenge due to higher applied field requirements. Improvements in the spatial resolution of PEEM through aberration correction optics have proved to be challenging, and will likely remain a very specialized technique. However, many prospects exist for improvements to the achievable time resolution through various bunch compression or slicing schemes. While laser-slicing techniques are likely too flux-limited to use with a PEEM, several storage rings are operating or commissioning low-alpha modes, in which the pulse length is shortened to several picoseconds, at the expense of some flux. At the Advanced Photon Source, the bunch deflection scheme of Zholents is under consideration, which would achieve a pulse duration of 1-2 ps. Finally, free electron lasers are becoming available in the soft x-ray regime (e.g., LCLS and FLASH), which offer the potential for sub-100 fs pulses, and single-shot imaging.
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