Chapter 1
Introduction

1.1 Layered Magnetic Structures

Not many fields in modern solid state physics have had such a dramatic impact on science and technology as thin film magnetism. More than 40 years ago, the coupling phenomena in multilayers were already predicted and described [1]. Ever since, a lot of effort has been dedicated on understanding the properties of magnetic films with thicknesses down to the few atom limit [2, 3]. The deposition of such films on top of supporting substrates bears the possibility to stabilize new materials by influencing their structure [4–8] and to study in detail the interaction between structural and magnetic properties [9, 10]. The latter were quite often found to deviate substantially from the properties known from the corresponding bulk magnetic materials. Finite size effects and the different coordination and environment of the atoms at the film's interfaces lead to deviating thermodynamic behavior [11–14], different spin [15–17] and orbital [18–21] magnetic moments, and different magnetic anisotropy energies [22–26]. More recently, novel ways to control spins, via pure spin currents, take into account non-collinear spin arrangements [27, 28], which are often amplified due to spin–orbit-driven interactions at such interfaces. Spin–orbit-driven interactions in layered systems of ferromagnetic and high-Z materials allow for an efficient electric current-induced domain wall motion [29]. Understanding the entanglement of the magnetic, electronic, and structural properties had, and still has, the attraction of finding ways to tailor magnetic properties in thin films by influencing parameters such as stress, strain, and electronic hybridization by the choice of the film and neighboring materials with respect to chemical composition and thickness.

1.1.1 Magnetoresistive Devices

Soon the focus in magnetic thin film research shifted from the investigation of single magnetic films to hybrid structures involving multilayered stacks of two or more magnetic layers, separated by non-magnetic spacer layers [30, 31]. The discovery of antiparallel coupling [32] and oscillatory interlayer coupling [33] in such multilayered systems paved the way for the controlled manipulation of the relative

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magnetization direction of different ferromagnetic layers. Ignited by the discovery of giant magnetoresistance (GMR) in magnetic multilayers at the end of the eighties [34, 35],[1] and the realization of its giant potential for applications [36, 37], systematic investigations of the effect were launched [33, 38, 39]. They were accompanied by fundamental studies of the magnetic properties of multilayered structures [40–42], which are important if one wants to provide suitable operating conditions for devices based on giant magnetoresistance. It took less than a decade from discovery to commercial exploitation of the effect. At the end of 1997 the first hard disk drives equipped with a read head of the new technology were sold to the customer. Nowadays, essentially the entire hard disk head production has been switched to multilayered magneto-resistive devices.

Figure 1.1 sketches the principle of such magnetic read head sensors, and some of the developments and visions that followed. Panel (a) presents a schematics of a current-in-plane GMR sensor, as it was used in hard disk read heads for a while. The electric conductivity of a stack of at least two ferromagnetic (FM) layers that are separated by non-magnetic but electrically conductive spacer layers can change significantly if the magnetization directions of the two magnetic layers are aligned more parallel or more antiparallel to each other. Technically, one of the magnetic layers is usually designed as a hard magnetic layer, which is only little affected by usual external magnetic fields, while the other layer, called the soft magnetic layer, is made to respond easily on changes in the applied magnetic field. The giant size of the effect allowed to detect small changes in magnetic stray fields from the hard disk as small deviations in the magnetic alignment of the soft layer, which in turn enabled disk manufacturers to decrease the amount of stray field above the disk needed to detect a single bit of information, and correspondingly to decrease its size. This has led to a change in the annual growth rate of storage density of magnetic hard disks, which before 1997 followed a rapid annual increase of 60 %, and since then has sped up to increase by an astonishing 100 % each year for a while.

A related effect, the tunnel magnetoresistance (TMR), is sketched in Fig. 1.1b. Although discovered before the GMR effect [43], it experienced a renaissance in the nineties, when it was discussed with respect to possible applications in magnetic data storage [44, 45]. The effect relies on the spin-polarized tunneling of electrons between two ferromagnetic layers [46–48] across a thin insulating barrier, typically an ultrathin oxide spacer layer. If one considers spin conservation in the tunnel process, the tunneling occurs between majority and majority states as well as between minority and minority states of the two FM layers, if their magnetization directions are parallel. If they are antiparallel, majority electrons from one FM layer tunnel into minority states of the other layer, and vice versa. In general, the density of the involved states and the tunnel probability will be different in these two cases, leading to tunnel magnetoresistance. Systematic fundamental investigations [49–54] eventually helped to push the tunnel magnetoresistance ratio at room temperature up to 220 % [55].

[1] The Nobel Prize in Physics 2007 was awarded to Albert Fert and Peter Grünberg for the discovery of Giant Magnetoresistance.

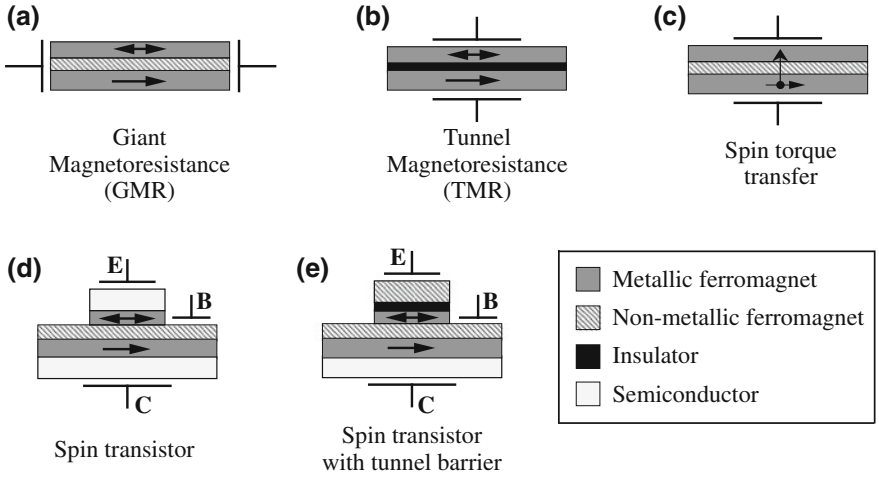

Fig. 1.1 Some modern concepts for devices based on thin film magnetism. **a** Giant magnetoresistance: Dependence of metallic conductivity in a stack of two or more ferromagnetic layers on the relative orientation of their magnetization directions. Soon after its discovery, this effect was applied in devices, for example, in read heads of hard disk drives. **b** Tunnel magnetoresistance: Dependence of tunnel current between two ferromagnetic layers across an insulating barrier layer on the relative orientation of the magnetization directions in the magnetic layers. This effect is, for example, currently applied in read heads of digital magnetic recording and will be used in future magnetic random access memories that are currently under development. **c** Spin torque transfer: Current-induced switching of the magnetization of a ferromagnetic layer in a nanostructured stack of two ferromagnetic layers by the torque exerted by the spin-polarized electrons on the magnetization of one of the layers. This is proposed as a means to switch the magnetization in magnetoresistive devices, and currently under intensive investigation. **d** Spin transistor: Three-terminal device in which the current between emitter (E) and collector (C) depends on the current between emitter and base (B) and, in addition, on the relative orientation of the two ferromagnetic layers. **e** Spin transistor with tunnel barrier: Three-terminal device like (**d**), in which in addition the emitter bias voltage can be used to tune the energy of electrons that are injected into the collector. The last two devices represent only two out of many approaches to the so-called field of spintronics, in which in future devices both the charge and spin of electrons are used for information processing

TMR elements have a higher resistance compared to GMR, which is desirable for certain applications, but the use of TMR requires the current to be run perpendicularly through the stack. In 2005, TMR-based spin valves have replaced GMR read elements in recording heads due to their higher magnetoresistance ratios and are in use up to now. Further downscaling, however, might enforce the use of current-perpendicular-to-plane GMR sensors in the near future as the high absolute resistance of TMR elements is a critical obstacle to downsizing such sensors [56]. Another possible application that is discussed in connection with the TMR effect is a magnetic random access memory (MRAM) (see Fig. 1.2). The idea is to have an array of small TMR elements like the one shown in Fig. 1.1b, which constitute the bits of the stored information, where "magnetization parallel" represents, for example, "0", and "magnetization antiparallel" stands for "1". The working principle is actually quite similar

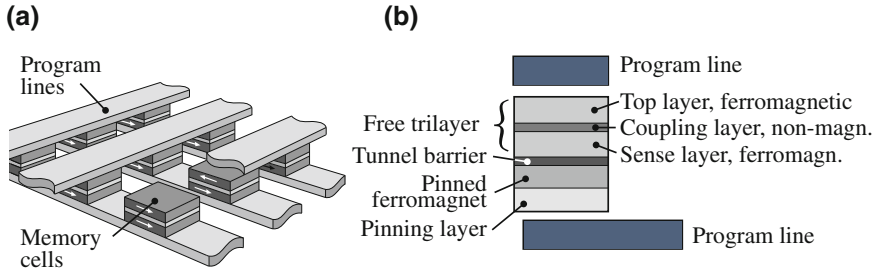

Fig. 1.2 Scheme of an MRAM device. **a** Simplified sketch of the arrangement of memory cells in an MRAM device. Reading of the information stored in the individual memory cells is achieved by measuring their electrical resistivity using the program lines and underlying diodes (not shown). **b** Detail of a memory cell. The tunnel magnetoresistance is determined by the relative orientation of the magnetizations in the sense layer and the pinned ferromagnetic layer (*small arrows* in **a**). The purpose of the pinning layer is to make the pinned ferromagnet insensitive to magnetic fields. The *top* and *coupling layers* are needed for a certain scheme for magnetization reversal in the so-called "toggle MRAM" [57]

to the magnetic core memories of the early days of computing. The most remarkable difference is obviously the size. MRAMs are being advertised as making the boot process of computers obsolete[2] when replacing conventional random access memory chips, because the magnetic information is retained after the power is shut off [58, 59]. First prototype MRAMs are commercially available for niche applications.

Reading the information in a TMR-MRAM means measuring the resistance through the stack. Addressing the resistance of a certain element in an array of trilayered stacks requires an integrated scheme of crossing word and bit lines with additional diodes underneath each stack in order to read the tunneling resistance of a particular element. The "0" and "1" states are then to be distinguished by some readout electronics. Technically even more demanding is the write process. It requires a very local and quite high magnetic field at the place of the element that is to be switched from "0" to "1" or vice versa, which means reversing the magnetization direction of the soft layer. Proposed writing schemes include a crossed arrangement of writing word and bit lines, through which relatively high currents can be run. The element to be switched is then the one at the crossing of the two lines under current, because only there the Oersted fields of the two lines add up to reach and exceed the critical field for magnetic switching. It is obvious that for the realization of a highly integrated MRAM device severe requirements with respect to the switching field distribution of the elements and the need to avoid cross talk between neighboring elements have to be met. Analytical tools to characterize the switching characteristics of these elements are mandatory for this.

[2] Note that the development of MRAM devices for applications in personal computers has to compete with the rapid progress in the development of semiconducting flash memories. It remains to be seen whether the use of MRAM will be restricted to niche applications, for instance where an insensitivity to strong radiation is required.

Another possibility of writing, i.e., of switching the magnetization in small elements, has been suggested after the discovery of current-induced effects on the magnetization. If a high current is vertically run through a multilayered stack as sketched in Fig. 1.1c, the electric current will be partially spin polarized after traversing the first magnetic layer. This spin polarized current exerts a torque on the magnetization of the other layer, and vice versa [60]. This can be used to reverse the magnetization of the soft layer, provided the current density is sufficiently high [61–66]. Typically 10^7–10^8 A/cm^2 are needed; this can be accomplished only if the area of the element is small enough.

1.1.2 Spintronics

Magnetoresistive effects and the generation of spin-polarized currents by ultrathin ferromagnetic layers have led to the vision of using not only the charge of the electrons for information processing, as in today's electronic circuits, but in addition also their spin [67–69]. This has been termed "spintronics". General fundamental questions regarding the efficiency of injecting spins into the FM layer (spin injection), the propagation of spin-polarized electrons (spin transport), the efficiency of detecting the spins (spin detection), and, generally, the inherent length scales of these processes (spin coherence length) are currently addressed in intensive fundamental studies. Recent reviews on spintronics can be found in [70] and [71]. A further emerging field of research, termed "spin caloritronics" [72], concentrates on the interplay between temperature and spin transport. This would allow to generate spin currents by temperature gradients, or use spin currents for heat transport.

Two possible logical elements that could be used as spin-sensitive logic devices are sketched in Fig. 1.1d, e. They are both three-terminal devices and have been termed "spin transistor" to emphasize the analogy to charge-based "conventional" electronics. Their working principle is as follows: A base current is run between emitter (E) and base (B). Due to the Schottky barrier formed at the semiconductor–metal interface, this leads to the injection of hot electrons into the metallic trilayer that forms the base [73]. These electrons are spin-polarized by the upper FM layer. Hot electrons travelling to the collector (C) are then spin-analyzed by the lower FM layer, so that the collector current changes its magnitude significantly if the magnetization configuration of the FM layers is switched from parallel to antiparallel. More than 300 % current enhancement at room temperature have been achieved [74]. Integrating an insulating barrier layer in such a three-terminal device, as shown in Fig. 1.1e, gives the experimenter the flexibility to operate the same device at different bias voltages, and thus to inject hot electrons at variable energy [75]. Optimization of a similar structure as the one depicted in Fig. 1.1e has led to a magnetoresistance effect of 3,400 % at room temperature [76]. The transfer ratio, that is the ratio between base and collector currents of such devices, is presently smaller than unity (between 10^{-6} and 10^{-4} [75, 77]), so that no amplification like in a real transistor is obtained. Still, the idea of programming or controlling logical operations by magnetism is

compelling and large efforts are dedicated to the development of more efficient spin injection into semiconductors [69, 78–86].

Another approach that is currently emerging involves the manipulation of magnetization in multiferroic systems by electrical fields. Multiferroic materials have coupled electric, magnetic, and structural order parameters that result in simultaneous ferroelectricity, ferromagnetism, and ferroelasticity [87–94]. With this coexistence, the magnetization can be influenced by an electric field and electrical polarization by a magnetic field, a property which is termed the "magnetoelectric effect". Aside from its fundamental importance, the mutual control of electric and magnetic properties is of significant interest for applications in magnetic storage media and spintronics, since the local application of electric fields to switch magnetic properties in nanoscale devices can be accomplished with much lower power dissipation than the application of magnetic fields. On a microscopic length scale it is expected that in ferroelectromagnets the coupled electric and magnetic ordering is accompanied by the formation of domains and domain walls, such that imaging techniques can contribute to the fundamental understanding of the interconnection of different order parameters.

1.1.3 Ferromagnetic–Antiferromagnetic Heterostructures

The coupling between an antiferromagnet and a ferromagnet at their interface is an issue that is important for the technical realization of magnetoresistive applications [38]. At the interface between ferro- and antiferromagnet the magnetic moments of both materials interact by exchange coupling. Studies of interfaces between a ferromagnet and non-magnetic adlayers have shown that the interaction with the ferromagnetic substrate may induce long-range ferromagnetic order in the adlayer, which by itself does not show ferromagnetic order [95–100]. The induced magnetization may be parallel or antiparallel to the magnetization of the ferromagnet, depending on the sign of the exchange interaction. This may lead to a ferromagnetic adlayer even for a material that in its bulk form is antiferromagnetic, as observed, e.g., for ultrathin Cr films on Fe [101–103]. In this case the long range order in the adlayer—be it an antiferromagnet or a non-magnetic metal in the bulk—is governed by the interaction with the ferromagnetic substrate and it may be expected to show the same temperature dependence. The temperature dependence of this induced magnetic order was in fact investigated for Mn on Ni and was found to be identical to that of the substrate [104]. Of course, as the antiferromagnetic layer thickness increases, the bulk antiferromagnetic state will prevail and each layer will show its own ordering temperature, approaching the bulk ordering temperatures for thick layers. It has been shown that the ordering temperature of the antiferromagnet may be influenced significantly by the presence of the ferromagnetic layer, as well as by its magnetization direction [105]. In addition, the exchange interaction at the interface can modify the magnetic behavior of the layered system in a characteristic way, which is known under the term "exchange bias".

Exchange bias was first observed in the 1950s on small Co particles covered with a Co oxide layer [106]. The hysteresis loop of these particles was shifted from being (inversion-) symmetric with respect to the magnetic field; instead, the inversion center of the hysteresis loop occurred at finite field, such as though some bias field was present in the sample, hence the term "exchange bias". The occurrence and magnitude of such a bias field depends on the history of the sample. To generate the bias field, the sample has to be cooled from above the Néel temperature—which has to be lower than the Curie temperature of the ferromagnet—in an external magnetic field. Obviously, this phenomenon is connected to the occurrence of magnetic moments in the antiferromagnet, which interact with those of the ferromagnet via exchange.

Exchange bias plays an important role for devices utilizing magnetoresistance effects or spin-dependent transport like the ones shown in Fig. 1.1. In such devices, one of the magnetic layers is required to have an increased coercive field for magnetization in a certain direction, which can be accomplished by exchange bias. Another example, the coupling of ferromagnetic nanoparticles to an antiferromagnetic material, was even suggested as a means of stabilizing the magnetic order at finite temperatures [107]. To take the full advantage of including antiferromagnetic/ferromagnetic interfaces into device structures, one needs to understand the underlying mechanisms governing the magnetic interaction at these interfaces.

Describing the exchange bias in terms of the magnetic moments of the antiferromagnet at the interface, which interact with the ferromagnetic moments by a typical exchange coupling [108], yields bias fields that are one to two orders of magnitude higher than observed experimentally. In the most simple model one assumes so-called uncompensated antiferromagnetic surfaces, i.e., surface orientations in which the atomic moments of the surface plane of the antiferromagnet add up to a non-vanishing net moment. The finite magnetization in the surface layer of the antiferromagnet interacts with that of the ferromagnet deposited on such a surface. The ferromagnet would then show a magnetization either parallel or opposite to the surface magnetization of the antiferromagnet, depending on the sign of the exchange interaction. However, it is not difficult to see why this simple picture is inadequate, because it does not take into account that any single-atomic step on the surface of the antiferromagnet will change the direction of the surface magnetization, which would enforce the formation of domains in the ferromagnet, or favor some other non-uniform state. Furthermore, exchange bias is also observed for interface orientations for which the antiferromagnet is not expected to have a net moment, the so-called compensated interfaces. This can not be accounted for by such a simple picture.

These aspects illustrate that the interaction between antiferromagnet and ferromagnet is rather complex. Therefore, the study of magnetic domains in such heterosystems [109, 110] provides an important input for modelling their behavior. By combining linear and circular magnetic dichroism in soft X-ray absorption, as will be explained in Sects. 2.7.1 and 2.7.3, layer-resolved magnetic imaging of both the magnetic domains in the ferromagnetic and in the antiferromagnetic layers is possible. Some examples will be presented in Sect. 4.4. The type of contrast—antiferromagnetic or ferromagnetic domains—is determined by the choice

of light polarization and by the choice of the absorption edges or the spectroscopic features used for imaging. The layer-resolved information allows one to correlate the occurrence of antiferromagnetic and ferromagnetic domains in the layered anti–/ferromagnetic system and to draw conclusions about the interaction between the layers.

1.1.4 Need for Layer-Resolved Information

Although some of the principles explained in Fig. 1.1 as well as the exchange bias effect are already employed in devices or are under development for applications, fundamental knowledge about the processes and the physics behind the important effects is still highly demanded. This is true for the static magnetic properties, for which the interplay with structural, electronic, and chemical properties can greatly help to achieve desirable behavior for the operation of devices, and this is also true for the dynamic magnetic behavior. The ever increasing need for data storage space driven by the ever progressing digitalization of all kind of media goes hand in hand with the demand for faster access to the stored information.[3]

All of the effects sketched in Fig. 1.1 have in common that the structures in which they are observed contain two or more magnetic layers within a multi-layered thin film structure, and that the magnetization of these layers has to be controlled independently. Furthermore, since these samples are often laterally structured or confined, nanoscale magnetic effects are becoming increasingly important. Apart from electronic transport properties, the understanding and the control of the magnetic coupling between different magnetic layers, also considering micromagnetic effects, is thus a major issue. To address the different layers separately and to obtain microscopic magnetic information about each layer in a simple way is thus crucial for the investigation of such structures.

Imaging of magnetic domains has essentially contributed at all stages to our present level of understanding of micromagnetic phenomena [111–114] in bulk materials as well as in magnetic films. Magnetic imaging techniques allow the most direct view on magnetic properties on a microscopic scale. The local magnetization direction as a function of geometric and material properties can be studied immediately in a straightforward way. Imaging the magnetic domain structures of multilayered magnetic systems separately for each layer with high spatial resolution, and to study the dynamics of the spin structure with high temporal resolution is therefore the outstanding challenge to any magnetic microscopy technique applied to all the systems described above.

A number of layer-resolving magnetic imaging techniques have evolved in recent years, which can be used to address the different magnetic layers in layered magnetic structures separately. They consequently offer substantial advantages for the

[3] While the length scales in devices approach the fundamental magnetic exchange length, the current time scales in magnetic applications are far from a fundamental limit (see Sect. 1.1.5).

microscopic investigation of layered magnetic structures compared to laterally or vertically averaging techniques. A review of these techniques is the objective of the present book, which intends to provide a comprehensive overview on the state-of-the-art imaging of magnetic domains in layered magnetic structures. It explains the fundamentals of the techniques and provides directions for potential users. Data and examples presented for each technique are (mostly) drawn from recent research, and are selected to illustrate the power for investigating complex multicomponent samples, such as ultrathin multilayers and nanostructures.

1.1.5 Need for Time Resolution

Beyond the static magnetic properties in nanoscale magnetic systems, the ultrafast spin dynamics in such systems is attracting more and more scientific and technological interest. The scientific goal is to understand magnetization dynamics on a fundamental time scale. Technologically the dramatic process of miniaturizing devices, e.g. in storage and sensor application, that has taken place over the last decade was by no means accompanied by a similar increase in speed of these devices. New concepts how to change the magnetization on a fast time scale need to be explored and are already being discussed, like the all-optical control of magnetization [115–117].

The general challenge to magnetic microscopies for studies of relevant layered systems is to reach simultaneously (i) nanometer length scales, (ii) ultrafast time scales, and (iii) depth or layer resolution. The time scales of interest in nanomagnetism cover several orders of magnitude from the micro- to nanosecond regime, where thermal processes and domain wall motion take place, to the nanosecond to picosecond regime, where precessional and relaxation processes occur, and down to the femtosecond regime, which is the fundamental time scale of exchange interaction, the strongest magnetic interaction.

The magnetization in a magnetic system can be changed, e.g., upon applying an external magnetic field. Since for the system this leads to a torque acting on the equilibrium magnetization, it will start a precessional motion that in turn relaxes after some time into the direction of the applied magnetic field. The same is true if a short current pulse exerts a spin transfer torque on the magnetization or if the effective magnetic field is temporarily changed by a short laser pulse. A phenomenological approach to describe the temporal development of the magnetization is provided by the Landau-Lifshitz-Gilbert equation

$$\frac{d\boldsymbol{J}}{dt} = \gamma\,[\boldsymbol{J} \times \boldsymbol{H}_{\mathbf{eff}}] - \frac{\alpha_{\mathrm{G}}}{J}\left[\boldsymbol{J} \times \frac{d\boldsymbol{J}}{dt}\right] \tag{1.1}$$

accounting for both the precession of the magnetization in an effective magnetic field H_{eff} with γ being the gyromagnetic ratio and $\boldsymbol{J}$ the vector of magnetic polarization, as well as for the relaxation and damping of the system. The latter is described by a phenomenological parameter α_{G} depending on the local geometry, anisotropy, and morphology. So far a complete microscopic understanding of relaxation processes

is lacking although it is of paramount importance for understanding microscopic dynamic properties of magnetization.

Spin dynamics can be imaged by time-resolved high-speed microscopy [118]. In such pump–probe experiments, the sample is excited by a continuously changing or pulsed periodic magnetic field, a current pulse, or a laser pulse. At certain time delays relative to the excitation, the magnetization is microscopically probed in a finite time window. Shifting the time delay of the probing window yields a series of time-resolved images of the magnetization process. Time resolution is either obtained by a gated high-speed video camera using a constant light source for illumination or by a pulsed light source and continuous detection. An ideal dynamic experiment should deliver a time-delayed series of single-shot images, each of them representing the momentary magnetic state of the sample during the evolution of the magnetization process within the same excitation cycle. Single-shot imaging, however, requires a sufficient amount of signal to be accumulated in the detector during the probing time in order to obtain a sufficient signal-to-noise ratio. Very bright light sources and highly sensitive image detectors are therefore necessary. Also, the repetition rate of the experiment has to be fast enough to provide adequately short time delays for in-cycle imaging. Both conditions are increasingly difficult to meet with rising excitation frequency or if the magnetization response is too fast after pulse-field excitation. If both, detector sensitivity and repetition rate of the experiment are limited, time-resolved microscopy has to be performed in a different way, known as stroboscopic imaging. In a strobed system, image acquisition is precisely synchronized to a periodic excitation, so that images are captured in the same time period of successive cycles and accumulated over many cycles until a sufficient signal-to-noise ratio is achieved. The time delay is then periodically shifted to temporarily scan along the magnetization process. This accumulation technique, however, requires repetitive magnetization processes during successive cycles.

Stroboscopic magnetic microscopy on magnetic films and multilayers can be realized in two ways: (i) By imaging techniques using visible light in laboratory-based wide-field or laser-scanning Kerr microscopes [118] (see Sect. 3.1). Time-resolution can reach the femtosecond range by applying corresponding lasers as light source. The lateral resolution in such systems is limited, though, to some 100 nm owing to the wavelength of visible light and so far time-resolved Kerr microscopy has never

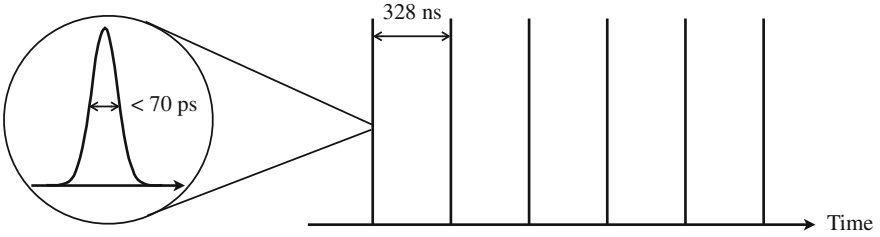

Fig. 1.3 Schematic of the time structure in two-bunch mode operation at the Advanced Light Source, Berkeley. The bunches are separated by 328 ns, while the width of each bunch determining the obtainable time resolution is $\approx$ 70 ps

been applied in a layer-resolved way although this would in principle be possible. (ii) By using X-ray illumination provided by a synchrotron radiation source. Here the lateral resolution is one or two orders of magnitude better, making synchrotron-based microscopy well suited for the investigation of micro- and nanostructures, and layer-resolved dynamic imaging has been realized already as will be demonstrated in Chaps. 4 and 5.

In recent years, the development of lab-based soft X-ray sources based on plasma sources and high harmonic generation has advanced significantly [119]. While these sources still can not compete with current synchrotron radiation facilities with respect to brightness, tunability to high photon energies, and polarization, their potential for the generation of femtosecond X-ray pulses with low jitter makes them very appealing for the study of dynamic processes [120, 121]. Although in principle imaginable, so far lab-based soft X-ray sources have not yet been used for layer-resolved magnetic imaging.

The X rays in a synchrotron radiation source are generated from electrons circulating the storage ring in short bunches at highly relativistic energies. At third generation synchrotron radiation sources the corresponding length of the X-ray pulses is in the sub-100-ps regime providing a time structure that is suitable for time-resolved studies of magnetism in layered structures. Since the intensity per bunch, however, is very low, such time-resolved studies have to be performed in a stroboscopic pump–probe scheme limiting them to fully reproducible processes. Synchrotron radiation facilities serving a large variety of user experiments at the same time can be operated in many different modes. Generally, there are hundreds of relatively small electron bunches stored in the ring and the ring cavities are running at typical RF frequencies, e.g. 500 MHz, therefore each bunch is separated by 2 ns. However, most synchrotron radiation sources offer few-bunch operation modes, where only one (single-bunch mode) or two (two-bunch mode) bunches are injected into the storage ring. These are very convenient for time-resolved studies if, e.g., the detected signal cannot be separated from other X-ray pulses by a gated detector or if studies require a longer gap between subsequent X-ray pulses, such as time-of-flight experiments. To illustrate this, the time structure of the two-bunch mode of operation of the ALS is sketched in Fig. 1.3. In this mode, there are only two bunches stored in the ring separated by 328 ns with a bunch width of about 70 ps. The single bunch mode of BESSY II offers 70 ps long bunches every 800 ns. In a dedicated operation mode with compressed bunch lengths, the so-called low-alpha mode, pulse widths below 5 ps are achieved [122].

At many synchrotron radiation sources also a hybrid mode is available in which one bunch that is different from all others, a so-called camshaft bunch, is injected from the accelerator into the storage ring. It has a higher intensity and is normally set into the ring pattern so that there is a large gap before and after this bunch. At the ALS, a 70 ps bunch is surrounded by breaks of 80 ns, while during the other 640 ns bunches appear every 2 ns. The camshaft can be used for time-resolved experiments

in which the other pulses can be blanked out,[4] serve as a marker, or can be used for special operations, such as the slicing mode offered at BESSY II in Berlin. In this slicing mode, femtosecond pulses are generated by modulating this bunch with a femtosecond laser pulse. This is an attempt to enable femtosecond dynamics experiments already with current synchrotron radiation sources. The drawback here is that the increased time resolution has to be paid with an enormous loss in intensity.

Next-generation sources using X-ray free electron laser systems will provide femtosecond pulses at very high intensity and therefore will enable studies of magnetism at femtosecond time scales that will not be restricted to pump–probe experiments, but will allow for single-shot experiments. Soft X-ray microscopies at such facilities hold the promise and the unique potential to cover both, fundamental length and time scales of magnetism, and provide elemental sensitivity, which is of paramount importance for studies of multicomponent systems.

There are several activities to combine X-ray-based layer-resolved magnetic imaging techniques with time resolution to study magnetization processes in multilayered magnetic systems on a microscopic length scale. This can be done by photoelectron emission microscopy (PEEM) or magnetic transmission X-ray microscopy in full-field (TXM) or scanning (STXM) mode by utilizing the time structure of synchrotron radiation sources as described above for soft X-ray pump–probe schemes. The results obtained so far are very encouraging since these time-resolved variants of the layer-resolved microscopic techniques overcome the limited spatial resolution of optical magnetic microscopy. Layer-resolved magnetic imaging by interference MOKE, on the other hand, has the prospect of combining the ultrafast laser pulses in the low femtosecond regime that are current state-of-the-art in the field of ultrafast laser technology with layer-resolved magnetic imaging. Some examples of time- and layer-resolved magnetic imaging using the three different techniques are presented in this book.

Chapters 4 and 5 close with some examples of time- and layer-resolved magnetic domain imaging. Using layer-resolved magnetic imaging for the investigation of fast magnetization reversal processes in multilayered or magnetically coupled systems is attracting more and more interest. One of the challenges for the further development of all kinds of magnetoresistive and spintronic devices will be to understand magnetization dynamics on a fundamental time scale in order to match the required increase in operation speed of applications.

[4] Current developments aim to kick the camshaft into a different orbit, which would allow single-bunch experiments in multibunch mode operation of the storage ring (pseudo single bunch). The user would be thus enabled to turn time resolution on and off for his (imaging) experiment.

1.2 Approaches to Layer-Resolved Magnetic Imaging

The approaches to layer-resolved magnetic imaging presented in this book are based on various magneto-optical microscopies, and thus make use of the interaction of light with the local magnetization of the specimen. There are, in principle, two interaction mechanisms that can be employed to achieve layer-selectivity:

- In the first, interference effects in layered structures are utilized to enhance or diminish the response from a certain depth of the structure.
- In the second, the different electronic properties of the different layers are utilized in order to selectively address a particular layer and probe its magnetic properties.

1.2.1 Interference-Based Approach

The interference-based approach to layer-resolved magnetic imaging uses conventional magneto-optical microscopy. Here the magnetic microstructure is visualized by polarized light in the *visible* frequency range in an optical polarization microscope. To be suited for microscopy, a magneto-optical effect must change the polarization state of light. This applies to four effects: the Faraday and Kerr effects (also known as circular birefringence effects), the Voigt effect (linear birefringence), and the gradient effect (a birefringence that depends on magnetization gradients). Common to all these effects is the generation of a magnetization-dependent magneto-optical light amplitude by illumination with plane polarized light. The phenomenology of the effects is introduced in Chap. 2, followed by a description of the microscopic techniques required to transfer them into a domain contrast in Sect. 3.1.

All the aforementioned effects have a certain depth sensitivity. This is trivial for the transmission effects (Faraday and transmission-Voigt effect), which are anyway applied on optically transparent materials such as magnetic garnet films. But also the reflection effects (Kerr, reflection-Voigt, and gradient effect) are depth-sensitive, providing information on the magnetization in a depth range of the order of the penetration depth of light. In case of metallic materials the *information depth* is about 20 nm. Compared to microscopy techniques using electrons or electron tunneling for imaging [like SEMPA (scanning electron microscopy with polarization analysis), SPLEEM (spin polarized low energy electron microscopy), SP-STM (spin-polarized scanning tunneling microscopy), or PEEM (photo-emission electron microscopy)], which exhibit depth sensitivities between one and a dozen atomic layers at the surface, conventional magneto-optical microscopy has in fact to be considered as a

depth-sensitive technique.[5] Within the penetration depth of light, phase differences between the magneto-optical contributions generated in different depths play a decisive role for the magneto-optical image. Usually these phase differences restrict the magneto-optical information depth to a stronger degree than absorption. In multilayer systems, however, the phase differences can be used to provide information on the magnetization of several ferromagnetic layers within the mentioned thickness range. Based on this "phase contrast", magneto-optical microscopy (notably Kerr microscopy) provides the possibility to image individual layers of a multilayer stack, even if the ferromagnetic layers are made of the *same material*. This is a fundamental difference to the second, electronic properties-based approach that relies on X-ray dichroism!

The physical basics of this information depth are described in Sect. 3.2, mainly for microscopy based on the Kerr effect, as this case has been most thoroughly investigated in the literature. Section 3.3 explains how the information depth of Kerr microscopy can be applied to obtain depth-*selective* information on magnetic domains in multilayer systems. Depth selectivity is achieved either by the phase adjustment of the different, interfering components of the reflected light wave by means of a rotatable compensator (like a quarter wave plate) or by tuning the photon energy or the angle of incidence of light. In this way the total Kerr signal from specific layers in a multilayer stack can be cancelled or enhanced, respectively, leading to images that represent the magnetic domain pattern at different depths of the sample or of different layers. Examples include the domain ground states of differently coupled epitaxial iron films that are interspaced by nonmagnetic layers as well as the switching of magnetic films which is dominated by stray-field interaction with domain walls in neighboring films of sandwich systems. Besides the Kerr effect, also the information depth of the magneto-optical Voigt and Gradient effects can be favorably applied to analyze coupling effects, especially in monocrystalline multilayers as reviewed in Sect. 3.4.

The interference-based technique thus represents a lab-based approach to layer-resolved magnetic microscopy without the need for polarized soft X rays that are generally only available at large-scale facilities such as synchrotron radiation laboratories.[6] In addition, the time resolution of conventional magneto-optical microscopy, if performed with laser illumination, exceeds that of present synchrotron radiation sources by about two orders of magnitude.

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[5] On the other hand, compared to integral techniques for the measurement of magnetization curves [like VSM (vibrating sample magnetometry) or SQUID (superconducting quantum interferometer device) magnetometry] the magneto-optical effects may be considered as surface-sensitive. As the Kerr effect is routinely used as a magnetization measuring tool particularly for ultrathin films, the acronym "SMOKE" [123–125] for "Surface magneto-optical Kerr effect" has been (somewhat misleadingly) invented for optical magnetometry based on the Kerr effect.

[6] There are current developments for lab-based soft X-ray sources, e.g., based on plasma sources and high harmonic generation. However, for the time being, they are not competitive to the brightness, tunability to high photon energies, and polarization of current synchrotron facilities.

An interesting, but different interference-based approach to depth-resolved imaging that is recently being explored relies on the interference of soft X rays. It has been demonstrated that by using a multilayer substrate as a mirror for soft X rays, a vertical X-ray standing wave can be created in a sample deposited on top [126–128]. By tuning the wavelength, the intensity maxima (or nodes) can be moved to different depths of the sample, which may be used for depth-resolved imaging [129, 130]. So far, layer-resolved magnetic imaging using that technique has not yet been realized.

1.2.2 Electronic Properties-Based Approach

For the electronic properties-based approach to layer-resolved magnetic information, the magnetic effects in resonant X-ray absorption spectroscopy are most convenient to use. These effects, termed magnetic dichroisms, occur for polarized X rays and are dominantly located in the vicinity of elemental absorption lines. Hence, any technique based on these effects exhibits an inherent elemental specificity. These lines correspond to the resonant excitation of core electrons by X rays into unoccupied electronic states. Particularly in the soft X-ray regime (200 eV–2 keV), i.e., for the L edges of transition metals and the M edges of rare-earth elements, the XMCD effects are very large (up to tens of percent), whereas in the hard X-ray regime (>2 keV) the XMCD effect is typically weak, generally in the sub-percent range. The use of these effects requires the possibility for tuning the X-ray photon energy. With the increasing availability of high-quality synchrotron radiation sources worldwide, however, this demand nowadays is not too difficult to be satisfied. The element selectivity of this spectroscopic contrast mechanism can be directly turned into layer selectivity provided that different magnetic layers contain different elements.

Magnetic dichroism techniques require polarized X rays, which, however, are easily available at synchrotron sources by either using the radiation emitted off-orbit at a bending magnet or by utilizing special insertion devices such as helical undulators. With regard to spatial resolution, X-ray based microscopies are advantageous. Compared to visible light microscopes, their shorter wavelength[7] pushes the diffraction-limited spatial resolution deep into the sub-10-nm regime. State-of-the-art X-ray microscopies have demonstrated spatial resolutions approaching the range of 10 nm and below [131].

There are two ways for detecting the X-ray absorption. One can either detect X-ray absorption by measuring the amount of emitted electrons at the sample surface, or by measuring the transmitted intensity of X rays after transfer through the sample. In laterally integrating experiments the former is usually termed “total electron yield” detection of X-ray absorption. The latter is a more direct way of measuring the absorption. The limited penetration depth of soft X rays of about 100–200 nm requires sufficiently thin specimens, but also matches the maximum thickness of interest in

[7] With λ[nm] $\approx 1.24/E$[keV] soft X rays have wavelengths of a few nanometers.

layered systems. Although the penetration depth for hard X rays reaches into the micrometer regime, the fact that the XMCD effects are very weak has so far prevented any significant utilization of hard X rays for magnetic X-ray microscopies.

In imaging experiments these two approaches are realized by combining resonant excitation by polarized X rays and existing X-ray microscopy techniques. A photoelectron emission microscope (PEEM) can be used to create a magnified image of the sample from the emitted electrons at the surface after photon absorption. Complementary to that, in transmission X-ray microscopy (TXM),[8] an image of the bulk properties of the sample is obtained from the different local transmission of X rays. This information is integrated along the photon beam, but laterally depends on the local sample magnetization direction if circularly polarized X rays are used and tuned to element specific absorption edges. These two basic approaches are explained and described in Chaps. 4 and 5.

Chapter 4 first describes the use of a photoelectron emission microscope for the imaging of magnetic domains in layered systems by using X-ray magnetic circular dichroism (XMCD). Presented examples include the study of magnetic interlayer exchange coupling in trilayered systems in which two ferromagnetic layers are separated by a non-magnetic spacer layer, the observation of transitions between collinear and non-collinear magnetic configurations in such trilayers, and the identification and quantification of micromagnetic coupling mechanisms mediated by the local magnetostatic stray fields from domain walls. A separate section describes layer-resolved imaging of antiferromagnetic materials in contact with ferromagnetic films by PEEM. The use of linear magnetic dichroism, as explained in Sect. 2.7.1, as a magnetic contrast mechanism for the imaging of domains in antiferromagnetic materials is explained. Examples of studies of the antiferromagnetic domain structure using linearly polarized synchrotron radiation are presented. By combining linear and circular dichroism, the correlation between the domain structure in the ferromagnet and in the antiferromagnet can be directly imaged and the coupling in combined ferromagnetic/antiferromagnetic systems can be studied.

In Chap. 5 the imaging of magnetic domains by magnetic transmission soft X-ray microscopy (M-TXM), which in contrast to the surface sensitivity of PEEM probes the volume magnetization in layered structures, is presented and reviewed. The strength of this technique is the high lateral resolution provided by Fresnel zone plates used as X-ray optical elements and the applicability of external magnetic fields during the image recording process, which allows detailed studies of magnetization reversal processes in layered systems. Layer sensitivity can be obtained both by the element-specific magnetic XMCD contrast mechanism and the different response of the magnetic domains to applied external fields. Presented examples include the layer-resolved imaging of magnetic domains in technologically relevant magnetically coupled systems and laterally patterned magnetic multilayers.

[8] The acronym TXM was chosen in analogy to TEM (transmission electron microscopy) to emphasize the fundamental difference of these two high-resolution microscopy techniques, i.e., electron versus X rays.

Before we turn to the different variants for layer-resolved magnetic imaging, Chap. 2 first introduces the basics of magneto-optic effects which affect the transmission and reflection in the regime of visible light as well as the absorption of soft X rays in the vicinity of elemental absorption edges. Magneto-optic effects in both these wavelength regimes constitute the basics of all the magnetic imaging techniques suitable for layer-resolved magnetic imaging discussed in this book.[9]

[9] A further technique, also capable of depth-selective imaging, is not treated in this book: By analyzing the backscattered electrons in a scanning electron microscope, a domain contrast (so-called Type II contrast) from depths of the order of 10 μm can be obtained, depending on the energy of the incident electrons. By tuning the electron energy, domain structures can be scanned at various depths [132, 133].

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