Journal of Electron Spectroscopy and Related Phenomena xxx (2013) xxx–xxx

Contents lists available at SciVerse ScienceDirect

Journal of Electron Spectroscopy and Related Phenomena

journal homepage: www.elsevier.com/locate/elspec

Magnetic imaging with full-field soft X-ray microscopies

Peter Fischer[a,*], Mi-Young Im[a], Chloe Baldasseroni[b], Catherine Bordel[c,d], Frances Hellman[c,d], Jong-Soo Lee[e], Charles S. Fadley[f,d]

[a] Center for X-ray Optics, Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, CA 94720, USA
[b] Department of Materials Science and Engineering, University of California Berkeley, Berkeley, CA 94720, USA
[c] Department of Physics, University of California Berkeley, Berkeley, CA 94720, USA
[d] Material Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94270, USA
[e] Department of Energy Systems Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 711-873, South Korea
[f] Department of Physics, University of California Davis, Davis, CA 95616, USA

ARTICLE INFO

Article history:
Available online xxx

Keywords:
X-ray magnetic dichroism
Soft X-ray spectromicroscopy
Spin dynamics
Fresnel zone plates
Mesoscale magnetism
Photoelectron microscopy
X-PEEM

ABSTRACT

Progress toward a fundamental understanding of magnetism continues to be of great scientific interest and high technological relevance. To control magnetization on the nanoscale, external magnetic fields and spin polarized currents are commonly used. In addition, novel concepts based on spin manipulation by electric fields or photons are emerging which benefit from advances in tailoring complex magnetic materials. Although the nanoscale is at the very origin of magnetic behavior, there is a new trend toward investigating mesoscale magnetic phenomena, thus adding complexity and functionality, both of which will become crucial for future magnetic devices.

Advanced analytical tools are thus needed for the characterization of magnetic properties spanning the nano- to the meso-scale. Imaging magnetic structures with high spatial and temporal resolution over a large field of view and in three dimensions is therefore a key challenge. A variety of spectromicroscopic techniques address this challenge by taking advantage of variable-polarization soft X-rays, thus enabling X-ray dichroism effects provide magnetic contrast. These techniques are also capable of quantifying in an element-, valence- and site-sensitive way the basic properties of ferro(i)- and antiferro-magnetic systems, such as spin and orbital moments, spin configurations from the nano- to the meso-scale and spin dynamics with sub-ns time resolution.

This paper reviews current achievements and outlines future trends with one of these spectromicroscopies, magnetic full field transmission soft X-ray microscopy (MTXM) using a few selected examples of recent research on nano- and meso-scale magnetic phenomena. The complementarity of MTXM to X-ray photoemission electron microscopy (X-PEEM) is also emphasized.

1. Introduction

Magnetic materials are the backbone of many key technologies, ranging from information and sensor technologies to transportation, power generation and conversion. A fundamental understanding of magnetic properties on the nanoscale is not only scientifically challenging, since it addresses the spins of correlated electrons, but is also of paramount importance for novel concepts and advanced technological applications [1,2]. A prototypic example for the intimate connection between scientific achievements and technological applications is the discovery of Giant Magnetoresistance (GMR), which not only contributed strongly to a profound understanding of the spin-dependent scattering of electrons in thin magnetic films, but had also a tremendous impact on the achievements in magnetic storage technologies immediately after its first discovery [3–5].

One of the primary goals in nanomagnetism research is to control spins on the nanoscale and there are several ways to do this. The field of spintronics exploits those mechanisms to develop new technological concepts based on the spin of the electron. Whereas the application of external magnetic fields (Oersted fields), which forces the magnetic moments to align parallel to the field direction, is still the basic concept to write information in a magnetic storage element, novel effects, using the local torque which is exerted by the electron spins in a spin-polarized current are intensely being studied, and have in fact recently been utilized in a commercial device [6,7]. Alternatively, in multiferroic materials, where multiple degrees of freedom, such as ferroelectricity and ferromagnetism, coexist, it has been shown that spins can be controlled by electric fields [8,9]. Unfortunately, there are not too many naturally occurring multiferroic systems, and therefore multiferroic materials are an intense topic in materials sciences, where advanced

* Corresponding author. Tel.: +1 510 486 7052.
E-mail address: PJFischer@lbl.gov (P. Fischer).

0368-2048/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.elspec.2013.03.012

synthesis techniques are utilized and developed to artificially fabricate such materials. Since multiferroic behavior is often induced by strain, "straintronics" has recently become a very active research area [10].

To understand magnetic properties on the nanoscale, advanced characterization tools are mandatory, and these should meet the following requirements:

- *spatial* resolution below 10 nm,
- *temporal* resolution down to the fsec regime,
- *chemical* and *magnetic* sensitivity with *elemental specificity*.

The first two requirements reflect the fundamental length and time scales of magnetism, while the last is a consequence of the fact that multicomponent materials can offer properties which differ from those of single-phase materials, thus motivating a "materials-by-design" approach. The first two requirements are related to the exchange interaction of individual spins, which is the strongest interaction in magnetic materials. Magnetic *exchange lengths*, which are largely determined by characteristic, i.e. material-specific, constants for exchange and anisotropy energies, are typically in the sub-10 nm range. The corresponding fundamental *time scale* for the exchange interaction, the exchange time, which can be derived from the Heisenberg uncertainty principle between time and energy, yields several tens of fsec. For comparison, typical time scales for the spin–orbit interaction have values in the psec regime, which are relevant for understanding, e.g. the dynamics of anisotropies. The time scales which govern, e.g. dipolar interactions or precessional motion of spins are in the nsec regime.

A multitude of magnetic characterization techniques has been developed in the past; however the utilization of soft X-ray spectromicroscopies offers a unique combination of relevant features and those are therefore among the most promising probes for nanoscale magnetic studies.

The uniqueness of soft X-rays for magnetism studies is intimately connected to their wavelength, photon energy and time structure, as well as their intensity and polarization characteristics [11]. With a wavelength spanning from about 5 nm to 0.5 nm, the diffraction limit, which corresponds to an inherent limit in spatial resolution or correlation length scales, is in the few nanometer regime. The corresponding photon energy between about 200 eV and 2 keV matches the inner core electron binding energies, which in turn give raise to an element-specific increase in X-ray absorption for those specific X-ray absorption edges. In particular, the L edges of 3d transition metals such as Fe, Co and Ni, as well as the M edges of rare earth elements happen to be in that regime. At current soft X-ray sources, such as synchrotron storage rings, the polarization of soft X-rays can be tuned from circular to elliptical and linear by dedicated sources, which allows using magnetic X-ray dichroism effects. They can be seen as the X-ray counterparts of well-known magneto-optical effects, such as the Kerr [12] and Faraday [13] effects, with the former referring to reflection, and the latter to absorption geometries). However, the soft X-ray analogs provide inherent elemental specificity and much larger magnetic cross-sections due to the strong spin–orbit coupling at the L_2 and L_3 (or M_4 and M_5) spin–orbit coupled X-ray absorption edges [14]. The application of magneto-optical sum rules to the X-ray magnetic circular dichroism (XMCD) effect further permits quantitatively extracting from XMCD spectroscopic data both the spin and orbital magnetic moments [15,16]. Lastly, synchrotron storage rings deliver burst of X-ray pulses, which provide an inherent time structure to those X-ray sources. Typically, at third generation sources, such as the Advanced Light Source (ALS) in Berkeley, these X-ray pulse lengths are in the <100 ps regime, but the next generation of X-ray sources, such as X-ray free electron lasers at the Linear Coherent Light Source (LCLS) in Stanford or the FLASH facility in Hamburg/Germany can now generate fsec short X-ray pulses with a peak brilliance which is orders of magnitude higher than what can be achieved today. For example, typical current X-ray fluxes are at about 10^{12} ph/s, whereas XFELs can produce about 10^{13} ph in a few fsec. Thus, exciting new opportunities for using soft X-rays in studies of magnetic materials are opening up.

While the understanding of nanoscale magnetic behavior is crucial and serves as the building block for novel magnetic devices, mesoscale systems have also recently received a significant interest. The mesoscale is not just defined as a length scale which bridges the nanoscale, i.e. the length scale of single atoms – to the microscopic range, where the magnetic properties act as a continuum, but it also adds complexity, stochasticity and functionality. It can be anticipated that effects which are related to those phenomena will see an increased relevance in future applications and will probably require new and extended theoretical models that bridge nanoscale and mesoscale behavior.

Complexity in magnetic materials will also become more important not only in multi-component combinatorial materials design approaches, such as the "materials genome initiative" [17], which aims to discover and tailor specific properties from basic principle calculations of novel materials, but also in devices extending into the third dimension, where the "simple" cross-talk in more common planar geometries will turn into corresponding volume effects. Related to this is the importance of interfaces, which play a crucial role in multi-component systems. The question of whether a specific magnetic process on the nano- or the meso-scale is fully deterministic or follows a *stochastic* behavior is not only scientifically fundamental, but moreover of utmost technological importance. Lastly, a steady-state characterization can only provide information about the static structure, but the *functionality*, e.g. of a magnetic device can only be understood if one is able to characterize the fast and ultrafast spin dynamics of a magnetic system, with high spatial resolution from the nano- to the meso-scale.

Imaging magnetic structures in three dimensions and in their corresponding fast and ultrafast spin dynamics in novel and advanced magnetic materials is thus not only a very appealing analytical approach [18], but it also provides detailed insight into magnetic behavior. Already, a variety of magnetic imaging techniques has been developed which permit studying static domain structures, i.e. the spin configuration in thermal equilibrium at highest spatial resolution. These techniques make use of various probes interacting with the magnetic material.

There are also optical microscopes using the magneto-optical Kerr effect (MOKE) [19], where a contrast is generated by the rotation of the polarization vector of the incoming light by a magnetic moment and where the utilization of ultrafast optical laser pulses has achieved a very fast (fsec) time resolution [20], albeit at moderate (diffraction limited) spatial resolution only.

There are electron microscopes, such as the Lorentz transmission electron microscope (TEM) [21], which utilizes the Lorentz force diverting the electrons as they propagate through the magnetic specimen, the Scanning Electron Microscope with subsequent Polarization Analysis of the electrons (SEMPA) [22], or the spin polarized low energy electron microscopy (SPLEEM) [23]. The X-ray photoemission electron microscopy (PEEM), which is one of the X-ray imaging techniques delineated in this review, can be seen as a hybrid between electron microscopies and X-ray microscopies, taking advantage of the limited escape depth of electrons in solid, which provides strong surface sensitivity [24].

There is also a variety of scanning probe microscopes, such as the Magnetic Force Microscope (MFM) [25], which senses the interaction of the stray field emanating from the sample onto its magnetic tip or the Spin Polarized Scanning Tunneling Microscope (SP-STM) [26], which detects the tunneling current between

Please cite this article in press as: P. Fischer, et al., J. Electron Spectrosc. Relat. Phenom. (2013), http://dx.doi.org/10.1016/j.elspec.2013.03.012