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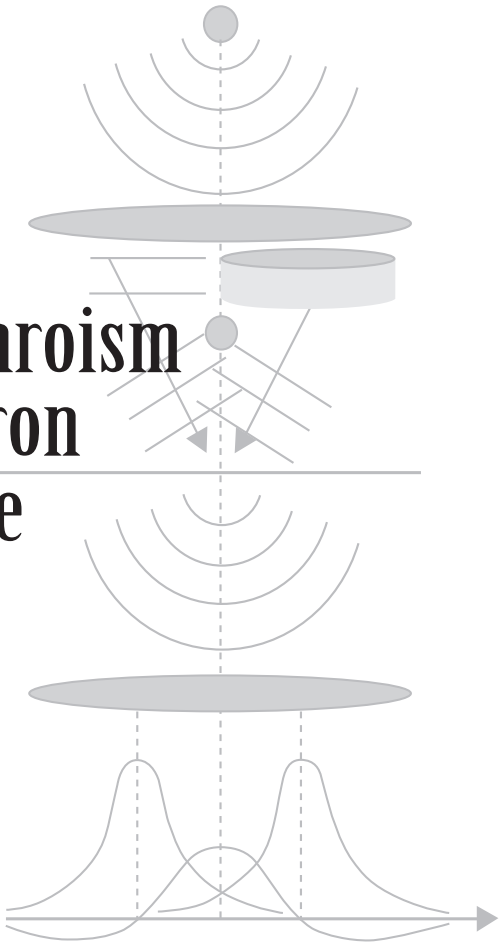

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Preface

Energy loss magnetic chiral dichroism (EMCD) is a rather new application that can detect element specific magnetic moments of solids in the electron microscope. The method is based on the analysis of ionisation edges in electron energy loss spectra (EELS), similar to X-ray magnetic circular dichroism (XMCD) in the synchrotron. EMCD was predicted in 2003 and experimentally verified in 2006. Compared to XMCD this was a very rapid development. Although ionisation edges in EELS (where the sensitivity to magnetic transitions is evident in retrospect) are well understood the discovery of EMCD came as a surprise to the community.

We know now from theory and simulations that *in principle* everything that can be done in the synchrotron is also feasible in an elecron microscope. The caveat lies in the *italics*; there are many obstacles in taking a dichroic energy loss spectrum that do not exist for synchrotron radiation. At the focus of these obstacles is the probe electron which reveals its difficult character here — it interacts stronger with matter, it is a Fermion, and it is more difficult to handle than a photon is. As a consequence, there are significant differences between EMCD and XMCD in spite of the similar theoretical description. In technical terms, these differences relate to the short extinction length for electron scattering on the crystal lattice, and to the high spatial resolution of electron microscopy. Recently, a resolution of about 1 nm in EMCD was demonstrated; the details are discussed in Chapter 12 of this monograph.

The intrinsic advantage of sub-nm resolution in electron microscopy allows new and exciting insights into the behaviour of electron spins and their interaction, a fact that is difficult to exploit because of the extremely faint signal. But EMCD is in rapid evolution. With new monochromators, aberration correctors and novel concepts for energy filters, pioneered by Harald Rose and others, the situation has improved. At the time of writing there are promising experiments under way in order to map spins of single atomic columns.

There has been so much progress in the last years that we (the authors of the following chapters and the publisher) considered the subject mature enough to present it to a broader audience. On second thoughts it became clear that the whole subject of symmetry breaking in EELS merits publication. It seemed appropriate to cover also aspects of anisotropy in EELS, both from an experimental and a theoretical point of view.

There are many open problems, such as the notoriously low L_2 signal which is not well understood. But now that the fundamental features of the method are well established it can be applied to real problems; most of them will be related to interfaces and nanoparticles. This is not so clear for another case of broken symmetry, namely linear dichroism in EELS; here, applications will probably focus

on the anisotropy of the density of states in larger systems (with examples given in Chapter 1), and on avoiding directional dependence of ionisation edges by use of the magic angle conditions, discussed in Chapter 7.

The book is organized as follows: the first three chapters are devoted to the basic principles of anisotropy and chirality, including an introduction to the XMCD technique. Chapters 4 to 8 cover the theory and numerics of the simulation of dichroic XMCD and EMCD spectra, as well as two aspects of symmetry breaking that are both theoretically demanding and important for the interpretation of experiments: the magic angle and the application of sum rules. Chapters 9 to 11 deal with the practical aspects of EMCD, such as the different techniques, data treatment, noise problems, the role of the crystal lattice, and typical applications. The final three chapters cover some newer aspects of EMCD that resulted from the refinement of experimental techniques and interpretation. For example, the problem of mapping magnetic moments in a lattice is intimately connected with Bragg scattering on that lattice. Is it then realistic to hope for spin maps on the atomic scale? Here, the chapter on magnetic X-ray holography, an elegant technique that holds promise for high resolution magnetic mapping, is well placed. Its penultimate position restores the broken symmetry with respect to the first part where XMCD was introduced.

I do hope that the reader will profit in several ways from this monograph — in conceiving new experiments in electron microscopy, in a better understanding of symmetry breaking in EELS, in combining EMCD with XMCD, and — maybe most importantly — in soliciting efforts to improve EMCD with the aim to make electron spins in the solid directly visible on the atomic scale.

P. Schattschneider
Vienna, March 2011

Foreword

It is more than seventy years since Siemens put the first commercial transmission electron microscope on the market. Over the decades, that first design has been improved by the inclusion of extra lenses and more coherent sources. Numerous accessories have joined the original instrument: stigmators, in-column and post-column energy analysers and, more recently, electron biprisms, monochromators and aberration correctors. New ways of using the microscope have emerged: Lorentz microscopy, the various holographic modes and of course the extensive family of diffraction techniques. Just occasionally, it is suddenly realised that, with no major modification, the electron microscope can detect physical phenomena that had seemed beyond its reach. The subject of this book is a striking example of such a revelation. Magnetic circular dichroism, which can be detected with X-rays, had been thought to be invisible to (unpolarized) electrons. But in 2003, Cécile Hébert and the editor of this book, Peter Schattschneider, suggested that this was not inevitable and a few years later, their prediction was confirmed experimentally. Circular dichroism can be detected in electron energy-loss spectra by a most ingenious manipulation of the experimental conditions. Full details of this and some related topics are provided in the chapters that follow and it is no part of my role to recapitulate them. What I do wish to emphasize is that the development of new techniques such as this requires an unusual (and often winning) combination of skills. Mastery of the theory of image formation and, in particular, of the theory of scattering in crystalline specimens is essential but not sufficient. To this must be added a good knowledge of the optics of the microsope and its accessories, notably, the interplay between the microscope lenses and the spectrometer. It is the marriage between these skills, not often united in a single individual, that leads to such developments. A comparable case is seen in ptychography. First suggested by the late Walter Hoppe, who was very familiar with TEM optics and the phase problem, and who was probably the first to realise that the STEM was well adapted for ptychography, it was subsequently made practical for the STEM by John Rodenburg, inspired by Richard Bates's instinct to Fourier transform everything in sight in difficult situations. Here again, we have an extremely original development, resulting from the union of the same two areas of expertise, instrumentation and image-formation theory.

This combination of skills, although vital for the discovery of new techniques, is fortunately not essential for the reader. Those who are closer to instrumental developments will enjoy the ingenious ways in which illumination conditions, specimen alignment and orientation relative to the spectrometer are juggled to yield signals strong enough to be detected while those primarily interested in

the materials science of magnetic specimens will appreciate the information that emerges and will no doubt attempt to apply the technique to their own specimens; and there is plenty of intriguing material for the theoreticians as well.

The experimental procedures have evolved considerably in the past few years and the whole subject is now mature enough to merit publication of this collection of essays on linear and circular magnetic dichroism. The authors of these contributions have collaborated in many of the published accounts with the result that the material is presented in a homogeneous fashion, doubtless largely thanks to the editor's influence.

I was asked to write a Foreword, not a "Forechapter", and it is therefore time for me to stop and to urge you to launch into this fascinating collection.

Peter Hawkes
Toulouse, May 2011

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Contents

Acronyms

AFM:	Atomic Force Microscope;
CBED:	Convergent Beam Electron Diffraction;
CCD:	Charge-Coupled Device;
CD:	Circular Dichroism;
DDSCS:	Double Differential Scattering Cross Section;
DFF:	Dynamic Form Factor;
DFT:	Density Functional Theory;
DoS:	Density of States;
EDX:	Energy Dispersive X-ray spectroscopy/analysis;
EELS:	Electron Energy Loss Spectrometry;
EFS:	Energy Filtered Series;
EFTEM:	Energy-Filtered Transmission Electron Microscopy;
ELNES:	Energy Loss Near Edge Structure;
EMCD:	Energy-loss Magnetic Chiral Dichroism;
ESD:	Energy Spectroscopic Diffraction;
FWHM:	Full Width at Half Maximum;
HWHM:	Half Width at Half Maximum;
HOLZ:	High Order Laue Zone;
LACDIF:	Large Angle Convergent DIFfraction;
LCC:	Laue Circle Center;
LCP:	Left-hand Circular Polarization;
LDoS:	Local(projected) DoS;
MCD:	Magnetic Circular Dichroism;
MLD:	Magnetic Linear Dichroism;
MOKE:	Magneto-Optic Kerr Effect;
MDFF:	Mixed Dynamic Form Factor;
PEEM:	Photo Emission Electron Microscopy;
RCP:	Right-hand Circular Polarization;
SAA:	Selected Area Aperture;
SEA:	Spectrometer Entrance Aperture;

SEM:	Scanning Electron Microscope;
SNR or S/N:	Signal-to-Noise Ratio;
STEM:	Scanning Transmission Electron Microscope;
STM:	Scanning Tunneling Microscope;
STXM:	Scanning Transmission X-ray Microscope;
TEM:	Transmission Electron Microscope;
XAS:	X-ray Absorption Spectrometry;
XMCD:	X-ray Magnetic Circular Dichroism;
XNCD:	X-ray Natural Circular Dichroism;
ZLP:	Zero Loss Peak.