# Contents

# Introduction

This dissertation reports on a systematic study of the thickness dependence of magnetic structures and phase transitions in helical-antiferromagnetic ultrathin lanthanide-metal films. The magnetic structures were investigated directly and in detail by means of scattering techniques. The advance of this study into the thickness range, where the magnetic properties are strongly altered by finite-size effects, became feasible with the development of resonant magnetic *soft* x-ray scattering and the construction of an ultra-high-vacuum compatible diffractometer, carried out in this work.

Lanthanide materials are particularly interesting for a basic understanding of electron correlation as well as magnetic properties of local-moment systems, but also due to their technological relevance. The interesting properties of the lanthanide metals and lanthanide-based materials arise from the open $4f$ shell that remains almost atomic like in the condensed phase. In the pure metals, the $4f$ moments at different lattice sites couple indirectly via polarization of the conduction electrons, i.e. by the so-called RKKY interaction, named after Ruderman, Kittel, Kasuya, and Yosida [1–3]. This interaction depends on details of the Fermi surface [4,5] and leads - together with anisotropies and magneto-elastic coupling - to a variety of exotic magnetic structures. For bulk materials, the magnetic structures and the influences of magnetic ordering on the crystalline lattice had been studied quite early by means of neutron and x-ray scattering [6]. Reliable experimental determinations of the electronic band structures, calculated about 30 years earlier, became only feasible with the availability of *in-situ* grown epitaxial films. These films provided atomically clean and single-crystalline surfaces, which are needed to explore band structures by surface sensitive photoelectron spectroscopy (PES) and inverse photoemission. Since then, lanthanide-metal films have developed into a rich playground in solid-state and surface physics. The observation of a magnetic exchange splitting of the valence states of ferromagnetic (FM) Gd by PES [7] initiated a wealth of investigations of the influence of collective magnetic order on valence states with different degrees of localization. The theoretically predicted behavior for localized and itinerant states [8, 9] was essentially found in a systematic study of magnetic exchange splittings in Gd, Tb, Dy, and Ho metal [10–13]. These elemental metals are particularly interesting since they share the same valence configuration and have essentially the same crystal structures allowing a systematic study of chemically and electronically similar systems, in which the local magnetic moments vary strongly.

Up to now, most theoretical and experimental studies of lanthanide-metal films and surfaces were devoted to the electronic structure and magnetic properties of Gd metal

[7–9, 14–20], which is a prototypical local-moment FM. Furthermore, most of the 'thin-film' experiments were done on comparably thick films, which have essentially bulk-like magnetic structures and magnetic ordering temperatures of the respective bulk materials. Thus, very little is known about the magnetic structures of these local-moment systems in the most interesting thickness range, where the magnetic properties are altered by finite-size effects as well as effects induced by the influence of surfaces and interfaces. One of the very few efforts in this field is the study of the thickness dependence of the magnetic ordering temperature of ultrathin Gd films [17]. In this study, a thickness-dependent reduced ordering temperature was found that follows a finite-size scaling law as predicted by theory [21–23], and as reported before for a number of FM transition metals [24–26]. Much less is known about finite-size effects in antiferromagnetic (AFM) systems and nothing at all about AFM lanthanide metals. While AFM CoO behaves in accordance with the scaling law found for FM systems [27], the spin-density wave of Cr in Cr/Fe multilayers behaves differently, following a modified, empirical scaling law [28, 29]. While there is at least some knowledge of the macroscopic magnetic properties of ultrathin films from these AFM systems, nothing is known about the details of their magnetic structures. This is due to the fact that the magnetic structures of films in this thickness range are barely accessible to conventional scattering techniques.

This dissertation is concerned with the magnetic structures and phase transitions in thin films of the heavy lanthanide metals Dy and Ho, in a thickness range where finite-size effects and effects induced by surfaces and interfaces are important. The magnetic structures were investigated by means of resonant magnetic x-ray scattering mainly in the soft x-ray region, which is a new technique that has been characterized in detail in this dissertation in order to obtain quantitative results.

*Chapter 1* introduces the electronic and magnetic properties of the heavy lanthanide metals Gd, Tb, Dy, and Ho relevant for the discussion of the results. Basics of x-ray scattering and magnetic x-ray scattering are discussed in *chapter 2* with a focus on scattering from thin films as well as magnetic scattering from Ho metal, which has served as a reference system in the literature. In *chapter 3*, experimental and technical details are described, such as sample preparation, the specially constructed experimental setups, and the synchrotron-radiation facilities, where the experimental results of this dissertation were obtained.

The main part of this work has essentially two goals: development of the method and insight into thin-film magnetism of AFM local moment systems. The methodic aspect are discussed in *chapter 4* and deal with the magnetic-scattering process in the soft x-ray region across the Ho $M_V$ absorption threshold. A model for data analysis, the optical parameters across the resonance, and special aspects of the resonant-scattering cross section will be presented in this chapter. Finally, the new method is compared to neutron scattering, which has been the standard method for magnetic-structure studies so far. In *chapter 5*, resonant magnetic soft x-ray scattering is applied to ultrathin Ho films, down to a thickness of 10 atomic layers. The influence of film thickness and boundary properties on the magnetic structure is discussed in detail. The main focus, however, is on the second-order phase transition in Ho metal films. A finite-size scaling law of the magnetic ordering

temperature of long-period AFM is determined and explained by mean-field calculations. By means of magnetic critical scattering observed over an unusually large temperature range, a dimensional crossover as a function of thickness and temperature was observed. A different and new application of magnetic scattering in the region of strong resonances at absorption thresholds is presented in *chapter 6*, where the magnetic depth profile during the first-order phase transition in Dy films is presented. This dissertation closes with a summary of the results.