Summary and Perspectives

The topic of this dissertation is magnetic structures and phase transitions of thin and ultrathin films of the heavy lanthanide metals Dy and Ho, studied by x-ray scattering, resonant x-ray scattering, and resonant magnetic x-ray scattering. By introducing new developments to resonant magnetic scattering in the soft x-ray region, it has become possible to study films in a hitherto not accessible thickness range, where interesting finite-size effects can be observed. The main achievements of this work are the design and construction of a fully ultra-high-vacuum-compatible two-circle diffractometer, the first detailed and quantitative characterization of the resonant-scattering process at the lanthanide M_V thresholds, and the application of the method to thin films. The phenomena studied include magnetic structures in thin Ho films, the thickness dependence of the magnetic ordering temperature in long-period antiferromagnets, critical phenomena in connection with the transition from three- to two-dimensional magnetism, and the characterization of depth-dependent magnetic profiles across a first-order ferro-to-antiferromagnetic phase transition in Dy films.

For the demanding experiments in the soft x-ray regime, a diffractometer was designed and constructed, which allows the *in situ* preparation of thin-film samples. The results of the present dissertation were obtained from lanthanide-metal films, epitaxially grown on a W(110) substrate crystal, as well as from films grown *ex situ* by molecular-beam epitaxy (MBE), embedded between Y layers. The films grown *in situ* revealed high quality with crystalline and magnetic coherence across the entire film and mosaic spreads similar to those of high-quality bulk single crystals. These films are structurally sufficiently simple to allow a detailed quantitative description of specular reflectivity scans. From the data obtained from the two types of films it was possible to obtain the optical parameters across the M_V resonance, necessary for any further quantitative studies, as well as the absolute values of the circular-dichroic and linear-dichroic components of the resonant scattering amplitude; the M_V resonance reveals scattering amplitudes up to $200\, r_0$, which is in the order of magnitude as predicted by Hannon *et al.* [Phys. Rev. Lett. **61**, 1245 (1988)], and which corresponds to an resonant enhancement of the magnetic-scattering cross section of more than seven orders of magnitude, opening up new opportunities for the study of thin films and dilute systems. A detailed comparison with magnetic neutron diffraction on identical samples demonstrated the enormous potential of the new method, with its much higher sensitivity.

Making use of the strong resonant enhancement of the magnetic-scattering cross section, the thickness dependence of the magnetic structure and magnetic ordering temperature of

Ho films was investigated down to a film thickness of ≈ 10 monolayers, which is of the order of the length of the magnetic period itself. It turned out that the magnetic structure of films thinner than about 15 monolayers is strongly influenced by their finite size, and by the properties of the boundary layers. While Ho films grown on W(110) tend - with decreasing film thickness - to a more FM alignment of the magnetic moments of neighboring layers, in the MBE-grown films, the influence of the surrounding Y leads to the tendency of a more AFM structure.

Although the development of the magnetic structure with decreasing film thickness is different for the two types of films, their thickness dependence of the magnetic ordering temperature is essentially the same, and is qualitatively different from the behavior of ferromagnets. While for the latter T_C follows a scaling law according to

$$\frac{T_C(\infty) - T_C(d)}{T_C(\infty)} = b \cdot d^{-\lambda},$$

T_N in the long-period antiferromagnet Ho is described by

$$\frac{T_N(\infty) - T_N(d)}{T_N(d)} = b' \cdot (d - d_0)^{-\lambda'},$$

including an offset thickness d_0, which can be understood as a minimum thickness to establish the AFM structure. This behavior appears to be a general property of long-period antiferromagnets. In collaboration with a theoretician, this originally phenomenological formula could be understood on the basis of mean-field calculations, which also establish that the offset thickness scales with the bulk magnetic period.

The high sensitivity of resonant magnetic scattering at the lanthanide M_V threshold to usually weak magnetic signals even permits critical-scattering studies above the magnetic ordering temperature in ultrathin Ho films. The obtained data give strong evidence that the films studied in the present work undergo a dimensional crossover in the region where the magnetic ordering temperature is significantly reduced and where a strong deviation of the magnetic structure from that of the bulk has been identified. This is indicated by an extended temperature interval of short-range magnetic correlations and by a qualitatively different behavior of the in-plane from the out-of-plane correlation length as expected for a 2D system. The thickness range in which the 3D $\rightarrow$ 2D transition occurs, lies between 16 and 11 monolayers, i.e. it appears again to be related to the magnetic period length.

Another and also new application of resonant magnetic soft x-ray scattering makes use of the dramatic change of the photon mean-free path at a strong resonances in the region of an absorption threshold. The resulting tunability of the probing depth was utilized to characterize the depth-dependent magnetization profile at the first-order phase transition between the FM and the helical AFM phase of Dy. This study was carried out for Dy films grown on W(110), a system characterized by two different interfaces on either side of the film. While an unusual non-abrupt transition from the FM to the helical AFM phase was observed with conventional x-rays, only the resonant magnetic soft x-ray study with tunable depth sensitivity revealed details of a temperature-dependent growth of AFM

domains. With the optical parameters across the Dy M_V absorption threshold determined separately, the magnetic depth profile could be established from temperature-dependent reflectivity scans recorded at various photon energies. It could be shown that the FM structure is stabilized at the W/Dy interface, while the helical phase develops gradually in the surface region, starting at a seed layer close to the surface.

Beyond the phenomena discussed in this dissertation, the results obtained have interesting implications for forthcoming studies. The strong magnetic sensitivity found in scattering experiments at the soft x-ray resonances opens the path for detailed studies of complex magnetic structures and correlations in thin films and dilute materials containing lanthanide metals. With a variety of different and exotic magnetic structures and properties even among the pure lanthanide elements, this dissertation might be the starting point for a systematic investigation of finite-size effects and for detailed studies of surface and interface influences. While the present work has focused on lanthanide systems, resonant soft x-ray scattering is not restricted to $4f$ elements. Strong dipole-allowed resonances in the soft x-ray regime exist also for the $3d$ and $5f$ transition elements, with wavelengths at the corresponding photon energies being well adapted to periodic structures in the nanometer range. Therefore, this method carries an extraordinary potential for the magnetic and structural characterization of artificial systems like multilayers, laterally structured materials, and other nanostructures in particular considering the tunable penetration depth of the x-rays.

Besides the huge resonant enhancement of the magnetic-scattering cross section and the strong variation of the photon penetration depth across resonances as used in the present dissertation, the spectroscopic information provides further detailed information on the electronic structure of the systems. Therefore, resonant scattering, particularly in the soft x-ray region, can be used as a unique tool for the study of ordering phenomena in connection with particular electronic states, like charge, and orbital ordering, as these are important for the understanding of highly-correlated electron systems.