Chapter 11

Summary and Outlook

The studies presented in this dissertation are devoted to three rather different physical effects observed at the surfaces of magnetic rare-earth materials by means of angle-resolved photoemission spectroscopy. Magnetic structure and Rashba spin-orbit interaction reflect the electronic ground state of a solid. Magnetic dichroism in photoemission, by contrast, is a final-state effect that becomes visible only upon optical excitations. The first part of this dissertation is devoted to studies of the magnetic structure at the Gd(0001) surface and its modification upon oxygen adsorption by mean of magnetic dichroism in photoemission. The second part deals with photoelectron diffraction in magnetic dichroism. The third part presents the first observation of the Rashba effect at magnetic rare-earth metal surfaces at the examples of gadolinium and terbium, and presents a way to strongly modify the Rashba splitting by formation of metal/metal-oxide interfaces.

From previous studies of the temperature dependence of the oxygen-induced exchange-split surface state on $p(1 \times 1)O/Gd(0001)$, interesting questions concerning the magnetic structure of this surface arose as well as about the role of the underling ferromagnetically ordered bulk. The results presented in this work give direct evidence that the magnetic structure of the surface monoxide layer formed on the Gd(0001) surface is not ferromagnetically ordered, in contrast to the underling bulk gadolinium. In combination, the studies of the MD effect and the Rashba effect, complemented by ab-initio band-structure calculations, lead to the conclusion that the spin polarization of the oxygen induced surface state is driven by the sub-surface ferromagnetic gadolinium layer.

The studies of the diffraction contribution to the magnetic linear dichroism (MLD) in 4f PE clearly demonstrate that diffraction can lead to very strong effects on the magnetic-dichroism PE spectra that can be dominant under certain conditions. The observations are explained by the different energy dependences of the excitation probabilities to dipole-allowed final states. Our semi-quantitative analysis implies that predominant one final-state channel excitation leads to a vanishing atomic part of MD, with the result that only diffraction dichroism remains. In these cases, the MD signal from the crystal surface reveals a strong modulation with the photoelectron emission angle, including a change in sign around the surface normal.

Using angle-resolved PE from the magnetically ordered surfaces of Gd(0001) and Tb(0001), as well as from their surface-monoxide interfaces, we demonstrated that the energy dispersion of the d_{z^2} surface state depends on the orientation of the spin-quantization axis of the electron relative to its propagation direction in the electric field that is provided by the surface potential. On clean metal surfaces, only the majority-spin surface state is occupied, requiring two subsequent experiments for the study of the spin dependence. In contrast to this, the exchange-split pair of surface states with opposite spin polarization on the monoxide surface allows to observe the spin dependence of the energy dispersion in a single experiment. Comparison of the experimental dispersion with first-principles DFT calculations including spin-orbit interaction gives evidence that this behavior is a direct manifestation of the Rashba effect. The fundamental difference between nonmagnetic and magnetic surfaces, concerning the manifestation of the Rashba effect, has been pointed out, and it was also shown why this effect is observable at some surfaces but negligible for others.

The present results represent the first observation of the Rashba effect on magnetic metal surfaces, as well as on the surfaces of the rare-earth metals. In view of the high atomic numbers of the lanthanides, the results of this work suggest the presence of sizeable Rashba splittings also on the surfaces of other metals of this series. As a result of this work, further studies of the Rashba effect at these surfaces are recommended that would certainly contribute to an improved understanding of the underling microscopic mechanism.

In connection with previous studies of surface states on Au(111) and W(110), the present results lead to the conclusion that the Rashba effect is a general surface and interface phenomenon. The strength of the effect is defined by the atomic contribution to the potential gradient experienced by the electronic states, by the shape of the electronic charge density distribution, and by the hybridization of the electronic states located at the surface/interface. We demonstrated that the strength of the Rashba effect can be changed by adsorbates, e.g. by oxygen adsorption, which leads to a strong modification of the near-surface electronic structure and in this way to an increased spin-orbit interaction.

The spin sensitivity of the Rashba effect suggests its possible utilization as a probe for the study of two-dimensional electronic spin structures. A combination of this information with magnetic dichroism data of PE experiments from core shells can provide a powerful tool for studies of surface magnetism.