6 Summary and outlook

A spin-resolved two-photon photoemission experiment is introduced with femtosecond time resolution, an energy resolution of about 70 meV and count rates in the order of 10^4 cps. This allowed us to determine spin-dependent electron dynamics, which will be presented in a thesis parallel to this one [*Schmidt*, 2007]. In the present thesis a study of image-potential states were employed to learn more about the fundamental properties of ferromagnetic materials in two aspects:

Image-potential states as a projector state

In the pump process, where electrons are excited from the initial states to the intermediate states, here the image-potential states, the spin is conserved. Therefore spin-resolved 2PPE via image-potential states directly relates to the spin polarization of the initial bulk states. Based on dipole selection rules the symmetry of the initial states is resolved changing the polarization of the pump pulse. The small bulk penetration of the intermediate image-potential states additionally provides a pronounced surface sensitivity. One may therefore state that the image-potential states are employed as projector states.

3 ML Fe/Cu(100) and 6 ML Co/Cu(100) served as first model systems. A significant signal from majority electrons with Δ_5 symmetry at the Fermi level reveals 3 ML Fe/Cu(100) as a weak ferromagnet whereas in the case of cobalt thin films the respective signal is weak and therefore suggests this system to be a strong ferromagnet. States with Δ_1 symmetry show spin-polarized features, which is in one case explicitly ascribed to a minority surface resonance by annealing experiments.

What is more, for the first time magnetic linear dichroism (MLD), i.e. the dependence of the intensity and the spin polarization on the magnetization direction, was observed in 2PPE. Both, spin- and time-resolved measurements allow an identification of the initial states according to their relativistic quantum numbers. We note that MLD only appears for *in-plane* magnetized samples, such as the ultrathin cobalt films in this work. One of the initial-state features which causes MLD can be related to the surface resonance mentioned above while the others are attributed to film-derived quantum-well states due to their thickness-dependent energy.

The spin polarization and dichroism in the 2PPE-signal, and in the low energy cut-off, which stems from direct photoemission are comparable. This demonstrates the surface sensitivity of photoelectron spectroscopy with very low photon energies, at least for the studied systems.

The results of this work shine new light on the surface-electronic structure close to the Fermi level. This forms a basis for band-structure calculations which may in the future explain the origin of the states and be used to predict novel physical properties of these ferromagnetic thin-film systems.

These dichroic measurements could be extended to systems where a strong hybridization of states with spin mixing is expected, preferably at the Fermi level with and technical relevance in ultrafast demagnetization processes and spintronics. A possible candidate is *in-plane* magnetized Ni/Cu(100).

One step beyond dichroism is the non-ferromagnetic correspondence, the linear spin-polarized effect (LSPE) [*Tamura and Feder*, 1991], which leads to a spin polarization of the excited electrons. This would provide the possibility to study spin-dependent electron dynamics in non-ferromagnetic systems, e.g. the spin dependence of the scattering potential when considering measurements for different electron wave vectors parallel to the surface.

Image-potential states as a sensor state

Not only the spin polarization within the image-potential states, which stems from the initial states but also the spin-dependent binding energy, observed in the n=1 and also the n=2 image-potential state, yields significant insight into magnetism-related properties. This has been demonstrated by measuring the temperature dependence of the n=1 state with respect to the difference in binding energy (exchange splitting), the spin polarization, the linewith and the energetic shift of the integrated peak maximum between spectra recorded with p- and s-polarized pump pulses. The latter two quantities allowed us explicitly to estimate the exchange splitting without spin resolution. The values obtained from the spin-resolved measurements suggest a much lower Curie temperature than those obtained from of the integrated measurements. This discrepancy can only be solved by considering two scenarios.

Firstly, one could think of a transformation of the film in a multi-domain state. Since the image-potential states are well defined within one domain the local exchange splitting still exists. By averaging over all domains any macroscopic spin polarization is lost. With increasing temperature, the magnetization in each domain decreases and therefore the exchange splitting is becoming smaller and finally zero at the real Curie temperature. This leads to the observed decrease of the linewidth of the single peak in the integrated spectra.

Secondly, one could also think of this multi-domain state as consisting of small spin blocks which are rapidly fluctuating in time and space in accordance to the fluctuating-band theory. A decrease of the exchange splitting could be explained by an increase of these fluctuations. Although a microscopic model of the magnetization behavior cannot be solely manifested, it is demonstrated that the image-potential states can be exploited as an ultrafast magnetization sensor.

In further experiments one may look for systems which provide an exchange splitting of the IPS which is directly observable in spin-integrated measurements. This would facilitate to determine the change in the exchange splitting with temperature.

To rule out a decay into a multi domain state at elevated temperatures one has to study systems with *in-plane* magnetization where this behavior is not expected. Such an experiment could finally provide fundamental insight into the existence of a short range magnetic order above the Curie temperature.

With respect to ultrafast demagnetization processes one could extend this experiment to a ”real”pump-probe experiment. A third laser pulse can be used as a perturbation which leads to a demagnetization whereas the former pump and probe pulse is used to measure the spin polarization and the exchange splitting of the image-potential states. These two values can be employed to quantify the transient magnetization and help to solve some of the yet unanswered question in ultrafast magnetization dynamics.