Chapter 5

Conclusion

The experimental observation of electron dynamics directly in the bulk can prove tricky, a fact to which the meagre amount of experimental data attest. Especially in a metal, where above all the dynamics occur on the few femtosecond timescale, it is difficult to differentiate between refilling and the various scattering processes an electron may encounter. However, years of research on image-potential states on noble metal surfaces have established this extraordinary class of electronic states as a perfect model system to study the elementary processes governing electron dynamics.

With these giant's shoulders to stand on, we approached the task of observing spin-dependent electron dynamics, our objective the experimental identification of microscopic scattering processes in ferromagnets. For this purpose we combined time-, angle- and energy-resolved bichromatic two-photon photoemission with spin-resolved electron detection. Though we unfortunately lose three to four orders of magnitude in intensity through diffraction in the spin detector, we could at least partially compensate these losses with our optimised setup.

We investigated thin iron and cobalt films grown on Cu(100). On both surfaces the lifetimes of image-potential-state electrons are spin dependent, more so on iron than on cobalt films. Based on the studies of the thin-film band structure by my colleague M. Pickel, smaller differences on iron as a weak ferromagnet with empty *d* states in both spin channels were expected. In a simple but generally rather useful approach, the decay rate of excited electrons in a metal is proportional to the density of unoccupied states. We would thus expect the spin dependence of lifetimes to be much more pronounced in the strong ferromagnet cobalt.

However, several theoretical studies suggest that for itinerant ferromagnets the excitation of collective modes, i.e. spin waves or magnons also has to be taken into account. This turned out to be especially true for iron, because on iron we found the signature of electron-magnon scattering in several instances, whereas such processes are much weaker on cobalt.

We were able to show in studies of the time-dependent lineshape of image-potential states that at the magnetic surface not only the lifetime is spin-dependent, but also the dephasing rate. Modelling time-resolved two-photon photoemission with optical Bloch equations yields that the linewidth of an intermediate state is solely determined by the experimental resolution and the dephasing rate after the pump pulse is over. A much larger linewidth of the minority-spin than the majority-spin $n = 1$ image-potential state at high pump-probe delay is evidence of population-conserving quasielastic scattering processes, that are stronger for minority-spin electrons. We interpreted these processes as scattering off 3 meV acoustic magnons on a femtosecond timescale. The lower lying majority-spin image-potential band allows electrons to scatter back and forth between the minority-spin band-minimum and the majority-spin band, while at the majority-spin band-minimum corresponding scattering partners are missing.

Spin-dependent scattering is likewise observed in resonant interband scattering. Electrons from the band minimum of both the minority- and majority-spin $n = 2$ image-potential state scatter quasi-elastically into the $n = 1$ image-potential band at finite parallel momentum. This leads to a biexponential decay of the $n = 1$ image-potential-state population, at large delay dominated by the long-living $n = 2$ -derived component.

Despite the high intensity of minority-spin electrons in the $n = 2$ image-potential state (which is related to the spin-dependent density of initial states), this resonant interband-component is significantly more pronounced in the majority-spin channel. We could safely exclude defect scattering as the source of this process. On the one hand, defect scattering, a process known to cause resonant interband scattering, revealed itself as not spin dependent on cobalt. On the other hand, if defect scattering on iron were spin dependent, it had to be much more effective on majority-spin electrons to create the measured effect. This would cause a larger majority-spin dephasing rate, in contradiction to our linewidth measurements. Here also the emission of magnons provides the most obvious explanation as a natural way for electrons to scatter from the minority-spin $n = 2$ into the majority-spin $n = 1$ image-potential band.

Apart from these quasielastic spin-dependent scattering processes with small energy transfer, we also examined energy relaxation processes within the image-potential bands. On cobalt, the increase of the decay rate with increasing energy above the image-potential-band minimum is only slightly spin dependent and comparable to the one on copper. This indicates that the increasing decay rate on cobalt can be attributed to the additional phase space available for decay, as in both systems phase space is gained in *sp* bands and the image-potential band only.

We found that intraband decay on iron is not only much stronger than on cobalt, but also highly spin dependent. In fact, the decay rate of minority-spin electrons increases twice as strong as the decay rate of majority-spin electrons. This factor of two can be most readily explained if electron-magnon scattering is included. Then minority-spin electrons may also scatter into the majority-spin bands via magnon emission and thus gain twice the phase space of their majority-spin counterparts. We emphasise that such a process can not occur directly, as Coulomb interaction does not mediate a spin flip (unless spin-orbit coupling is strong). Magnon emission must take place through the exchange of a minority-spin with a majority-spin electron from the bulk and multiple interaction of the final-state electron-hole pair. We thus interpret the strongly spin-dependent intraband scattering on iron in terms of magnon-enhanced exchange scattering between opposite spin-bands, consistent with theoretical predictions that spin-flip processes accompanied by spin-wave emission contribute significantly to the decay of minority-spin electrons in iron.

In conclusion we have observed ultrafast electron-magnon scattering of image-potential-state electrons in three different regions of energy and momentum transfer directly in the time domain. Though the signature of magnons can be found in SPEELS [176, 171] and high-resolution photoemission spectra [148], such measurements can not give information about the time required for the emission of a magnon, the latter experiment may not even imply the emission of a magnon at all [79].

The spatial separation of image-potential states from the substrate causes their electron's dynamics to occur on a slower timescale, though the dynamics will still be dominated by the underlying bulk electrons. Because in the image-potential states electron-electron and electron-magnon scattering rates are comparable, the same can be assumed for bulk electrons. This means that even low-energy magnons can be emitted within a few femtoseconds.

Contrary to popular belief, the energy transferred in a scattering process of an electron with a quasiparticle, e.g. a phonon or a magnon, does *not* determine the scattering rate of this process through the uncertainty relation. It is the interaction strength, which is responsible for the timescale of scattering processes and the uncertainty relation enters insofar as on timescales smaller than the oscillation period of the exchanged quasiparticle, energy conservation can be (temporarily) violated [62, 17, 101].

These arguments and our time-resolved measurements lend support to the conclusion that electron-magnon scattering is ultrafast. Thus magnon emission does not constitute a bottleneck for the speed of magnetic switching and may very well be a microscopic process involved in femtomagnetism.