Abstract

It is known from thermodynamics that a ferromagnet heated above its Curie temperature loses its macroscopic magnetization. However, thermodynamics does not describe the velocity of this process. It was long believed that the time it takes for the magnetization of transition ferromagnets to react on a sudden temperature increase is limited by the weak spin-orbit interaction to the 100 ps time scale. Therefore, it was a big surprise when Beaurepaire et al. showed in 1996 that the magnetization of nickel can be reduced on the sub-picosecond time scale when heated by a femtosecond laser pulse. Since this discovery, the mechanisms responsible for ultrafast demagnetization are still highly debated. The most often employed technique to access the ultrafast magneto dynamics is the magneto optical Kerr effect. However, it has been argued that the strong laser excitation might induce state blocking effects such that the observed ultrafast demagnetization is mimicked by optical artifacts. Furthermore, most optical methods rely on the spin-orbit interaction which has been shown to be altered during the demagnetization. In this thesis, a method is described which overcomes these problems by measuring the time evolution of the spin polarization of the cascade electrons by time and spin resolved photoemission. Photon pulses (40/180 eV), generated by the free electron laser (FEL) in Hamburg, provide enough energy to release electrons from everywhere within the valance band. These photo-electrons represent, therefore, an average magnetic signal of the sample. With this technique, we could show that the demagnetization time in iron is indeed ultrafast and comparable with values measured by the magneto optical Kerr effect. In addition, it was found that the FEL pulses alone can reduce the measured spin polarization. This effect is, however, not caused by an FEL induced demagnetization. The reason is space charge – the vast number of electrons released start to repel the low energetic but highly spin polarized electrons and reduce therewith the measured spin polarization. This finding will help to design forthcoming photoemission experiments at free electron laser based sources. Photoemission has in general a low electron yield. This complicates spin resolved photoemission experiments as the widely employed Mott spin polarimeters have additionally a low detection efficiency. In collaboration with Prof. Schönhense's Group (Mainz, Germany), a high efficiency spin polarimeter was developed. An iridium crystal acts as a spin selective electron mirror and allows, in conjunction with a hemispherical energy analyzer, the simultaneous measurement of the spin polarization, the energy, and the electron's emission angels. This multidimensional detection scheme has the potential to allow for future time-, angle-, energy-, and spin-resolved photoemission experiments. In search for the mechanisms at play responsible for the ultrafast demagnetization, two important correlations were discovered and are presented in this thesis: The demagnetization time scales with magnetic film thickness and electrical resistivity. Based on these two observations, a semi-classical thermodynamical model is developed explaining ultrafast demagnetization as a spin transport effect with localized spin-flips at the ferromagnet/substrate interface. We propose the spin currents to be caused by a chemical potential gradient generated by the absorption of the infrared pump pulse. The localized spin-flips solve the dilemma of how spin transport can also induce ultrafast demagnetization in a ferromagnet deposited on an insulator. The model gives realistic values of the demagnetization time and amplitude for the transition ferromagnets without any fit parameter. Moreover, the model predicts that laser pulses of equal energy, but longer pulse length, should result in a reduced demagnetization and that the ultrafast demagnetization should be slower for a hot sample than for a cold one. Both propositions are shown to hold experimentally for the case of nickel. Therewith, the experiments provide evidence that the Elliott-Yafet spin-flip mechanism is unlikely to be the main cause for the ultrafast demagnetization in nickel. The proposed model provides an intuitive mechanism for ultrafast demagnetization based on spintronics fundamentals.