Chapter 1

Introduction

Today the magnetic storage industry achieves bit write times in the low nanosecond range [172]. Conventionally the magnetisation of a magnetic particle in a magnetic storage device is reversed by applying a magnetic field antiparallel to the magnetisation. The write time is then limited by the speed of domain wall nucleation and propagation [34].
With precessional switching, where the torque from a perpendicular applied field rotates the magnetisation coherently (exchange interaction forcing the spins to remain aligned), switching times are (theoretically) limited only by the strength and timescale of the magnetic field pulse [110]. However, there seems to be an upper boundary in the magnetic field strength above which the dynamic behaviour of the magnetisation becomes unpredictable [6]. So far coherent magnetisation reversal has been experimentally demonstrated to work within $100 - 300$ picoseconds [155, 64].
In 1996, Beaurepaire and coworkers challenged this timescale by demagnetising a thin nickel film with a strong laser pulse in less than half a picosecond [10]. Similar investigations [77, 154, 83, 76, 67, 97, 68, 143] confirmed that ultrafast demagnetisation of thin ferromagnetic films upon optical excitation occurs within a few hundred femtoseconds. The interpretation of the dynamics observed in these experiments in terms of magnetisation dynamics remains controversial [98, 84, 102]. There are, however, several arguments in favour of a true ultrafast demagnetisation. First, various experimental techniques were employed, among which are magneto-optical Kerr effect (MOKE), spin-resolved photoemission, magneto-optic second harmonic generation (SHG) and microwave emission, all reporting a similar timescale of $100 - 300$ femtoseconds. Second, also a complete quenching of the magnetisation could be achieved and observed with two different experimental methods [9, 38]. Third, most alternative interpretations like bleaching effects could be ruled

out with careful repetition and analysis of the experiments and the dynamical behaviour observed with magneto-optical effects is independent of geometry (Kerr or Faraday) and independent of probe pulse wavelength [20]. Last, after the quenching of the magnetisation reaches its maximum, the magnetisation slowly recovers as the electronic system is cooled via electron-phonon scattering and eventually a Landau-Lifshitz-Gilbert-like precession of the magnetisation vector around its equilibrium direction can be observed, which supports the claim of observing true magnetisation dynamics [95].
For a physical rather than phenomenological interpretation of the quenching of the magnetisation within a few hundred femtoseconds we are confronted with a far greater problem. Whenever the magnetisation changes direction or magnitude conservation of angular momentum must be observed. While spin ($\vec{S}_e$) and orbital ($\vec{L}_e$) momentum of the electronic system yield the magnetic moment of the system according to

$$\vec{\mu} = \mu_B(\vec{L}_e + g\vec{S}_e) \ , \tag{1.1}$$

they contribute to the total angular momentum of the system as

$$\vec{J} = \vec{L}_e + \vec{S}_e + \vec{L}_{\text{lattice}} + \vec{L}_{\text{photon}} \ , \tag{1.2}$$

together with the contributions of lattice and photon field. Ultimately the spin system will equilibrate with the lattice via electron-phonon scattering. But this takes place on the much longer electron-lattice relaxation time, typically in the picosecond range.
Several attempts at an explanation on the microscopic level have been published since the first experiment in 1996. A complete transfer of spin to orbital momentum could reduce the magnetic moment to no less than one half. The successful total quenching of the magnetic moment mentioned above and no indication of spin-to-orbit transfer in preliminary magnetic circular dichroism experiments [46] seem to rule out momentum transfer. An exchange of angular momentum with the photon field constitutes an allowed channel for a fast reduction of $\vec{M}$, provided that circularly polarised light is absorbed or emitted or linearly polarised light is absorbed in a dichroic medium [191, 190]. Though photons in the THz range are indeed emitted during demagnetisation, they are essentially linearly polarised [11]. Additional estimates based on the laser parameters in the experiments [96] and time-resolved photoemission data [143] conclude that the contribution of the laser field is one to two orders of magnitude too small.
Due to the timescale of the demagnetisation and the explanations already excluded, the remaining interpretations concentrate on processes *within* the

electronic system. Spin-flip scattering of the excited electrons accompanied by Stoner excitations or magnon (spin wave) emission was suggested as a fast channel for the reduction of $\vec{M}$. While it is true that the excitation of a magnon lowers the magnetisation by $2\mu_B$, magnon emission (without the participation of the lattice) occurs through the reversal of a minority- into a majority-spin electron, which corresponds to an increase of $\vec{M}$ by exactly $2\mu_B$. Hence these processes do not change the total magnetic moment of the electronic system [90].

Koopmans and coworkers propose that a demagnetisation time shorter than electron-phonon equilibration is achieved if an Elliott-Yafet type of spin-flip probability is included for each electron-phonon scattering event [94, 95, 37]. Though the probability required in their model calculation exceeds the average spin-flip probability in $3d$-transition metals by two orders of magnitude, band structure effects may increase this value significantly [53]. It must be said that despite "unifying" theories repeatedly being put forth, so far no satisfactory explanation of the phenomenon has been found.

We believe that the key to understand this phenomenon lies in understanding the spin-dependent scattering processes in a magnet. For this we require a well-defined model system with accessible electron dynamics, which is sensitive to the magnetisation and which allows us to distinguish between different scattering processes.

Unlike in a metal, in a semiconductor the strength of the main scattering processes, i.e. defect scattering, phonon scattering and electron-electron scattering can often be tuned by the doping density, the applied field, the growth mode and the temperature over many orders of magnitude. At the metallic surface, however, a certain level of control over the influence of phonon and defect scattering or the interaction with the bulk electrons can be achieved. Much work has already been done on metal surfaces, both theoretically and experimentally, for the identification of scattering processes, summarised only recently in [48].

One comparatively successful approach to study electron dynamics in a metal is two-photon photoemission of image-potential states. Image-potential states reside several Ångstrom in front of the surface. They interact only weakly with the bulk electrons and therefore experience the dynamics in "slow motion", very beneficial for experimental accessibility. Additionally, no transport phenomena or spin-filter effects during transition through the surface obscure the experimental data like in photoemission of bulk states. Thorough studies of the scattering behaviour of image-potential state electrons in front of noble metal surfaces provide a reliable basis for our experiment

(e.g. [58]).
In front of a ferromagnet, the exchange interaction allows us to observe the magnetisation of the sample. The spin-resolution opens an entirely new playground, namely the investigation of spin-dependent scattering processes in ferromagnets.
It is therefore the aim of this work to contribute to the knowledge about spin-dependent electron dynamics in a d-band ferromagnet, with the ultimate goal in mind to help uncover the microscopic processes responsible for femtomagnetism.