1 Introduction

The continuous and increased demand for a higher density data storage and a faster data access (read/write) in the computer industry triggered a tremendous development of the field of magnetization dynamics over the past decade [1, 2]. In magnetic recording the digital information is stored in the form of small magnetized regions or "bits", whose opposite magnetization or spin orientations correspond to "1" and "0" values. In order to write a bit an external magnetic field can be applied reversing the magnetization of the magnetic bit. Thus, the fundamental questions which the scientific community and the industry have to face and to address are: how one can switch the magnetization of a certain specimen and how fast one can do it.

In this context it is not surprising that "smaller and faster" is the logo that drives the actual development in magnetically-based computer industry. Regarding the "smaller" term, the areal density of nowadays magnetic devices is approaching fast the fundamental limit of superparamagnetism where the thermal energy k_BT is large enough to produce fluctuations of the overall spin orientation of the magnetic nanoparticle used to store a bit of information *i.e.* leading to a loss of information. For the "faster" term there is still place for improvement since the actual switching speed of the state-of-the-art devices lies in the low limit of GHz range *i.e.* it takes about several ns (1 ns = 10^{-9} seconds).

The manipulation of magnetization in direction and magnitude can be achieved mainly by employing stroboscopic or pump-probe techniques. Here the *pump* pulse, that perturbs the magnetization ground state, can be either a pulsed magnetic field or a femtosecond (1 fs = 10^{-15} seconds) laser pulse whereas a weaker, time-delayed, laser beam can be used as a *probe* to detect the subsequent transient magnetization dynamics.

Using pulsed magnetic fields, the magnetization can be reversed from "0" to "1" (or viceversa) state due to the torque exerted by the effective magnetic field B_{eff}, which leads to a precession of the magnetization vector along B_{eff} during the reversal event. With this method the magnetization orientation is reversed with no change in its magnitude at switching rates in the GHz frequency range [3, 4]. However, precessional switching driven by pulsed magnetic fields exhibits the fundamental limit of half a precession period *i.e.* on a time scale of few hundred of picoseconds [2]. Using femtosecond laser pulses higher frequency spin waves could be obtained (up to 140 GHz) by excitation of transient magnetic anisotropy fields [5, 6] exploiting the dependence of magnetic anisotropy on the laser-induced temperature rise. Only very recently Kimel *et al.* [7] have been shown that employing the photo-magnetic inverse Faraday effect a coherent magnetization dynamics up to a frequency of 400 GHz could be achieved[1]. Nevertheless, these time scales are orders of magnitude slower than the femtosecond laser pulses which could be used, in an ideal case, for data writing.

[1] In the same study a non-thermal excitation of the magnetic system has been observed within the laser pulse duration of 200 fs.

In addition to coherent magnetization dynamics, the utilization of femtosecond laser pulses determine the decrease in magnitude or even quenching of magnetization on a subpicosecond time scale [8, 9] due to laser-induced excitations. A new and exciting research field is emerging namely magnetism on the femtosecond time range: *the femtomagnetism*. In short, upon absorption of a femtosecond laser pulse the photoexcitation energy resides in the electronic system that leads to an increase of its temperature T_e. Subsequently, the energy is redistributed to another degrees of freedom of the system namely to the lattice and spin subsystems. Depending on the characteristics of the system the demagnetization occurs by a direct electron-spin coupling or via electron-lattice-spin interaction. As can be already noticed, in order to understand the laser-induced demagnetization it is crucial to identify the elementary processes (electron-lattice, electron-spin, lattice-spin interactions) that lead to demagnetization of the system, the time-scales on which they evolve and their relative weight with respect to the demagnetization process.

This thesis reports about *laser-induced coherent spin dynamics* at a frequency of several THz, that is at least one order of magnitude faster than previously reported, as well as *ultrafast demagnetization dynamics* on a *femtosecond time scale* ($<$ 100 fs). The system under study is the rare-earth gadolinium (Gd) metal, that is considered [10] as a prototype of a Heisenberg ferromagnet due to the localization of the electrons responsible for the ferromagnetic ordering. The coherent and incoherent magnetization dynamics are investigated in the present work by means of a time-resolved nonlinear magneto-optical technique *i.e.* magnetization-induced second-harmonic generation (MSHG). The MSHG is the tool of choice in this work since is very reliable in investigating magnetization dynamics owing to its high sensitivity to magnetization effects at surfaces and interfaces of centrosymmetric media [11], where the inversion symmetry is broken.

The usual approach in studying ultrafast magnetization dynamics was to investigate $3d$ ferromagnetic materials *i.e.* Fe, Co, Ni. In these materials the magnetism is governed by the *delocalized* or itinerant $3d$ electrons and one can have a direct access to the magnetic moments by means of a ultrafast laser with photon energies in the visible range. Employing various time-resolved experimental techniques such as magneto-optical Kerr effect (MOKE) [8], magnetization-induced second-harmonic generation (MSHG) [9] and two-photon photoemission (2PPE) [12] the early stages of magnetization dynamics have been accessed. The conclusion was that *laser-induced demagnetization evolves on a subpicosecond time scale ($<$ 500 fs) but the elementary processes which are responsible are still not unambiguously identified*. In this respect, a crucial point is the conservation of angular momentum during the demagnetization process on the ultrafast time scale. Hence, in spite of the improved level of understanding gained from the manifold of performed investigations, there still exists a *"terra incognita"* at the early times in the photoinduced spin dynamics.

In the present thesis a different approach in accessing ultrafast magnetization dynamics has been followed:

- The *femtosecond* laser-induced demagnetization dynamics of the *localized magnetic moment* ferromagnet gadolinium, has been investigated. In gadolinium the ferromagnetic ordering is determined by an indirect exchange interaction (RKKY) among the localized $4f$ magnetic moments mediated by the $5d6s$ conduction electrons, with

the former ones providing ≈90% of the total magnetic moment of Gd. Due to their binding energies (9 eV below Fermi level) the $4f$ electrons cannot be accessed by lasers with photon energies in the visible range. The optical excitation and detection of magnetization dynamics evolve via the $5d6s$ conduction electrons. In the view of these facts there are a few questions that arise. Does the localization and indirect coupling of the $4f$ electrons slow down the laser-induced demagnetization? What are the spin-scattering elementary processes responsible for demagnetization? What are their characteristic time scales? How does this compare with the photoinduced magnetization dynamics on the itinerant ferromagnets?

- The optical-induced magnetization behavior is addressed by a systematic approach involving the MSHG tool that measures the spin polarization in the surface region. An independent 2PPE measurement performed in our lab by M. Lisowski and P. Loukakos under similar conditions on the same system provide us with information regarding the transient behavior of the exchange-split electronic structure. Based on the information acquired from both investigations we can propose a demagnetization mechanism [13] that challenges the common believe in the community regarding laser-induced magnetization dynamics, which settles the limits at around 0.3 ps [14]. The suggested mechanism is based on the spin-flip scattering of hot electrons with emission/absorption of magnons, a process that is acting within the first 100 fs after excitation and is mediated by a strong electron-magnon coupling present in Gd. The picture of the proposed demagnetization scenario can well be extended on the time scale of non-equilibrium electrons *i.e.* before electronic thermalization and therefore this thesis is the one of the first reports that shed some light on the femtomagnetism *"terra incognita"*.

Beside the induced electron and spin dynamics, the excitation of a solid with femtosecond laser pulses can trigger also coherent lattice dynamics. These coherent optical phonons, with typical frequencies in the range of a few THz, have been extensively studied on semimetals, semiconductors and high-temperature superconductors employing time-resolved optical techniques [15, 16]. Such coherent optical phonons were not expected to occur in metals due to the effective screening of the spatial electron redistribution, that takes place on a time scale equal with the inverse of the plasma frequency (attoseconds time scale). This thesis represents the first study that reports the observation of *coherent optical phonons on a metal surface*. The coherent lattice vibration is triggered by the excitation of the exchange-split surface state components on Gd(0001) surface, and is quasi-instantaneously (within our time resolution of 50 fs) coupled to a coherent spin excitation, both oscillating at a common frequency of 3 THz [17]. Here the time-resolved linear and nonlinear optical techniques are employed, that, according to their sensitivity, allow us to disentangle between surface and bulk dynamics. The coupled phonon-magnon quasiparticle is remarkable since it brings to light a *novel phonon-magnon coupling mechanism* that is based not on the usual spin-orbit coupling, but on the modulation of the exchange interaction strength J by lattice vibration. Moreover, the coupled phonon-magnon mode constitutes itself as a model system for studying the excitation and subsequent relaxation of quasiparticles in ferromagnetic Gd. By varying several experimental parameters like

temperature, laser wavelength and sample morphology a wealth of information could be retrieved regarding the excitation, interaction and the lifetime of the involved quasiparticles: electrons, phonons and magnons.

In addition to the above presented investigations, the physics of *propagating acoustic phonons* at GHz frequencies is addressed in the second investigated system in this work namely epitaxial yttrium films on a tungsten substrate Y(0001)/W(110). The concept of travelling acoustic phonons, the so-called *phonon echo*, is very useful in investigation and diagnostics of materials *e.g.* presence of defects since it has a non-contact, noninvasive and nondestructive character. Also a precise evaluation of thickness and sound velocity can be performed using the laser ultrasonics technique. The laser ultrasonics denote the pump-probe technique which monitors the modulations in the refractive index of the system produced by the sudden distortion of the lattice upon absorption of a laser pulse, that travels as a GHz frequency acoustic pulse. Upon strong laser excitation the presence of a long lived coherent acoustic phonon mode bouncing back and forth in the Y film is revealed. This is ascribed to the high optical absorption of the film, to the high acoustic impedance of the Y/W interface and to the smoothness in the interface region. An accurate estimation of sound velocity in the epitaxial Y thin films prepared under ultra-high vacuum (UHV) conditions could be done.

The present thesis is structured as follows. In chapter 2, the electronic structure and magnetism of Gd as well as the general features of laser-induced phenomena such as electron, lattice and spin dynamics are introduced. In chapter 3 the basics of nonlinear optics are introduced emphasizing the role of nonlinear magneto-optics and in particular of magnetization-induced second harmonic generation that is the tool of choice in this work. The experimental details concerning the ultra-high vacuum system used to grow epitaxial rare-earth thin films as well as the femtosecond laser system that is the light source for ultrafast spectroscopy, are described in chapter 4. Chapter 5 presents the laser-induced phenomena on Gd(0001)/W(110) system comprising incoherent and coherent magnetization dynamics as well as coherent electron and lattice dynamics. Coherent lattice dynamics in GHz regime on Y(0001)/W(110) are detailed in chapter 6. The conclusions and outlook of the present work are given in chapter 7.