7 Summary

This thesis presents the femtosecond laser-induced electron, lattice and spin dynamics measured on two representative rare-earth systems: the ferromagnetic Gd(0001) and the paramagnetic Y(0001) metals. The main issues addressed in this work are:

- the investigation of the excitation mechanism, the interaction and the relaxation of the coupled coherent lattice and spin excitations that evolve at THz frequencies on the Gd(0001) surface.
- identification and separation of the elementary spin scattering processes that are responsible for the ultrafast demagnetization dynamics, following the excitation with a femtosecond laser pulse, on the localized magnetic moment ferromagnet Gadolinium.
- study of the physics of the propagating acoustic phonons in Y(0001) thin films and retrieval of the elastic and photoelastic characteristics of the system.

The employed investigation tools are the time-resolved linear reflectivity and second-harmonic generation, which offer simultaneously complementary information about the bulk and surface/interface dynamics, respectively. In the following the obtained results are detailed.

Coherent lattice and spin dynamics

The optical excitation of the exchange-split surface state of Gd(0001) triggers simultaneously the coherent vibrational dynamics of the lattice and spin subsystem in the surface region at a frequency of 3 THz. The electron/lattice and spin dynamics could be separated owing to the symmetry of the even and odd SH fields with respect to magnetization reversal. The coherent optical phonon measured by the even SH field represents the vibration of the topmost atomic layer against the underlying bulk along the normal direction to the surface. This periodic variation of the interlayer distance between two adjacent hcp planes modulates the exchange interaction J between neighbor atoms that gives rise to an oscillatory motion of the magnetic moments at the same frequency with the lattice vibration. Thus, we show the possibility of the spin system to follow the lattice coherently at THz frequencies. The coupled coherent phonon-magnon mode represents a new type of magneto-elastic interaction, that is mediated not by the spin-orbit coupling but by the dynamical variation of the exchange interaction. The excitation mechanism of the lattice vibration has been identified as being DECP(displacive excitation of coherent phonons)-like from the initial cosine phase of the oscillations. The charge-driven character of the excitation mechanism has been proved in wavelength-dependent measurements. Independent support comes from the time-resolved photoemission measurements that show an oscillatory motion of the majority surface state binding energy with the same frequency as the lattice vibration. Upon elevating the temperature of the system we could identify the scattering with electrons as the dominant relaxation pathway of the coherent phonon. Contributions coming from the phonon-phonon and phonon-magnon scattering, are also considered.

Femtosecond demagnetization dynamics

Another goal of this project is to study the femtosecond laser-induced demagnetization on the local magnetic moment ferromagnet gadolinium, and to identify and disentangle the elementary processes responsible for the ultrafast loss of magnetization. For this purpose the dynamics of the spin polarization and of the exchange splitting of the Gd(0001) surface state following the excitation with a femtosecond laser pulse have been studied. Employing pump-probe MSHG the spin polarization of the Gd(0001) surface state is measured, which exhibits a sudden drop within the laser pulse duration of 50 fs. An independent time-resolved photoemission study [13], performed under similar conditions, shows a constant exchange splitting of the surface state. From these observations has been concluded that on Gd(0001) the spin polarization does not follow the exchange splitting dynamics and that on ultrafast time scales the spin-mixing behavior dominates. The time scale on which the demagnetization occurs is at least comparable if not smaller than observed for the itinerant ferromagnets. The ultrafast decrease of spin polarization can be explained by the quasi-elastic spin-flip scattering of the hot electrons among spin-mixed states. The angular momentum conservation is fulfilled by emission or absorption of magnons, that can involve also the $4f$ moments.

Phonon echo dynamics

Epitaxially grown thin films of Y(0001) on W(110) substrate have been investigated with time-resolved linear reflectivity and second-harmonic generation. Upon pump pulse excitation, in the transient LR signal sharp peaks with alternating polarity have been observed, that appear at regular time intervals. This has been interpreted as the signature of propagating acoustic phonons at sound velocities within the film, the so called phonon echo phenomenon. The phonon echo signature is long lived on Y(0001)/W(110) system due to the high optical absorption at 800 nm, the high acoustic impedance of the film/substrate interface and due to a presumably smooth film/substrate interface. The phonon echo is excited by the sudden temperature increase produced by the absorption of the femtosecond laser pulse, which leads to a lattice expansion that launches a strain pulse. The values for the film thickness, sound velocity and optical penetration depth could be retrieved from the phonon echo data. The central frequencies of the travelling strain pulse ranges from 70 GHz to 200 GHz, that can be varied as a function of the film thickness. Using the thermoelastic model we could compute the shape of the first phonon echo, and from here we deduced the values of the photoelastic constants of the Y(0001) thin film.

Spin-polarized adsorbates investigated with magnetization induced sum-frequency generation

Furthermore a pilot experiment has been developed within the framework of this thesis, in which the CO molecules adsorbed on the ferromagnetic thin films of Ni on Cu(100) substrate are investigated. The goal of this project is to explore the capabilities of the new technique of magnetization-induced sum-frequency generation (MSFG) on a model system characterized by a spin reorientation transition Ni/Cu(100). For this purpose a new UHV system has been constructed and the magnetic characterization of Ni/Cu(100) films as well as the TDS (thermal desorption spectroscopy) measurements of CO on Ni/Cu(100) have been performed.

Future developments

Based on the insight developed in the present study there are are several points that can be investigated in the future. With regard to the coherent lattice and spin dynamics, two similar rare-earth systems Tb(0001) and Dy(0001) are very interesting candidates since exhibit an exchange-split surface state and the same valence electronic structure. Also they have a similar phonon spectrum but with a higher magnetic anisotropy and implicitly an active spin-orbit coupling. Moreover, the phonon and magnon dispersion curves do not overlap anymore, the magnons having lower energies. Thus, the excitation and the coupling mechanism of the coherent phonons and magnons can be tested. Also these two systems are suitable to investigate how the spin-orbit coupling and the higher magnetic anisotropy influence the femtosecond magnetization dynamics observed on Gd(0001). For both the coherent and the incoherent spin dynamics, time-resolved investigation with femtosecond light sources that can reach the localized $4f$ moments (9 eV binding energy), like UV and XUV free-electron laser or the generation of higher-harmonics in gas cells, would be very valuable. Also a time-resolved XMCD experiment, that could disentangle the orbital and spin contribution to the total magnetic moment, might clarify whether the $4f$ are involved in the coherent and incoherent spin phenomena encountered on the Gd(0001) surface.