Summary

The subject of this thesis concerns time-resolved spectroscopy on ultrashort time scales. The properties we consider in this thesis are the dynamics of an electron gas and the time resolved propagation of light through a disordered strongly scattering medium.

To be able to study the fore mentioned effects, one has to know how the light interacts with the system which is studied. This interaction is described by the response function of the system. In case of interferometers, one usually calls the response function the transfer function. This transfer function describes the scattering of light from the interferometer and is a complex valued function. In chapter 2 we discuss transfer functions from a general point of view. Transfer functions should obey a Kramers-Kronig relation, enabling one to calculate the real part form the imaginary part of the transfer function and vice versa. To see whether a Kramers-Kronig relation exist for a transfer function, it is sufficient for a transfer function to be analytic. In chapter 2 we study the analyticity of transfer functions of interferometers. It is shown that if one uses the Fourier-Laplace transform to calculate the transfer function in the frequency domain, the tranfer function is analytic and a Kramers-Kronig relation can be formulated. In the literature it was recognized that problems can occur if one tries to formulate Kramers-Kronig relations between amplitude and phase. We show that if we formulate the proper transfer function this transfer function obeys a modified amplitude-phase relation.

In chapter 3, we present a novel method to determine the full transfer function of a system, based on measuring the interferometric cross-correlation function between a pulse propagated through the system and an undistorted reference pulse. The method is demonstrated experimentally by measuring the complex-valued transfer function of an etalon in transmission. The experimentally obtained results are compared with the calculated transfer function and are found to be in excellent agreement. Furthermore we show that the time resolution of the method is about one order of magnitude better than the pulse duration of the pulses used in the experiments.

The extremely high time-resolution of the novel method makes it sufficient to study the time-resolved propagation through multiple scattering media as we describe in chapter 4. Important parameters which characterize the multiple scattering medium are the thickness of the medium L and the average

path length between two scattering events ℓ. The coefficient L/ℓ defines the optical thickness of the multiple scattering medium. An exact description of the propagation of light through these multiple scattering media is to difficult. Therefore, in this field it is customary to describe the transport of light through multiple scattering media with a diffusion equation, which is shown to be correct for optically thick media ($L/\ell >> 1$). For optically thin media it is expected that diffusion breaks down. Up to now optical experiments to study the time-resolved propagation through multiple scattering media in the visible regime of the electromagnetic spectrum have been performed for optically thick media ($L/\ell >> 1$), the reason being that no methods were available having time-resolution sufficient to study optically thin media. The novel method we introduced in chapter 3 has a time-resolution sufficient to study optically thin samples ($L/\ell \sim 1$). The experiments we performed mainly concern the question whether or not diffusion theory is also applicable for these optically thin media and we show that the diffusion approximation clearly breaks down for the dynamic measurements. We also performed stationary measurements. In these measurements the time integrated total transmission is recorded. Contrary to the dynamic measurements these results could be described with standard diffusion theory. We tried to interpret the experimentally obtained results with theory that goes beyond the diffusion description.

In chapter 5 we describe experiments performed to study the electronic relaxation processes in gold. Usually these type of experiments are performed using an optical pump-probe measurement. Commonly used theories to describe the relaxation dynamics of the electrons assumed that after excitation of electrons, electron-electron collisions maintain an equilibrium within the electron gas. However, strong evidence that this equilibrium was not established was obtained using pump-probe experiments. Due to the finite time-resolution of an optical pump-probe experiment, the initial relaxation processes, which is the interval where we expect to see large deviations from the equilibrium of the electron gas, are difficult to resolve. Using the new method we were able to study this time interval. Efficient excitation of electrons and sensitive detection of the changing reflectivity properties of the gold film were achieved by employing the surface-plasmon polariton resonance at the metal interface. Essential to be able to analyze this type of measurement is a consistent modelling of the influence of the excitation of hot electrons by the leading part of the pulse on the relectivity properties during the trailing part. We performed numerical calculations to model these phenomena, showing that due

to the nonthermal distribution of the electron gas the resonance frequency of the surface-plasmon polariton shifts. The experimentally obtained results did not show a convincing shift of the resonance.

In the final chapter we present measurements of the scattering properties of particles on a substrate. The particles we study have a diameter in the order of the wavelength of light. The scattering properties of these particles in a homogeneous surrounding are described by exact Mie theory. However, in a non-homogeneous surrounding no theory is available. There are two reasons to study the scattering properties of a particle near a surface. In the first place, the measurements clearly show that the scattering properties of light by a particle are influenced by its surrounding. A proper formulation of the scattering properties of a particle in a finite geometry may serve as an explanation for the break down of diffusion for thin samples as described in chapter 4. Due to the finite geometry a large influence on the scattering properties of the particles is expected. The other reason is the analogy with near field optics. The system of a particle near a surface is a system similar to a STM probe tip near a surface. We performed two types of experiments: a measurement on a single scatterer on a substrate and a measurement on a lattice of scatterers. The measurements showed strong deviations from Mie theory.

For further reading we refer to the following articles:

Chapter 2: Rik H.J. Kop, Pedro de Vries, Rudolf Sprik and Ad Lagendijk, Kramers-Kronig relations for an interferometer, Optics Communications **138**, page 118-129 (1997).

Chapter 3: Rik H.J. Kop and Rudolf Sprik, Phase-Sensitive interferometry with ultrashort optical pulses, Review of Scientific Instruments **66**, page 5459-5463 (1995).

Chapter 4: Rik H.J. Kop, Pedro de Vries, Rudolf Sprik and Ad Lagendijk, Observation of Anomalous Transport of Strongly Scattered Light in Thin Disordered Slabs, Physical Review Letters **79**, page 4369-4372 (1997).