Synopsis

The interaction of electromagnetic radiation with matter is the basis of most of the processes observed in nature, and this phenomenon has always aroused the curiosity of mankind. Under ordinary circumstances the optical properties of a material remain unchanged, regardless of the intensity of light falling on it. However, this is not true at higher intensities where the optical properties become a function of the input light intensity. With the development of science and technology newer sources of light as well as novel forms of matter have emerged, enabling a deeper study of light-matter interaction. Advances in laser technology during the last two decades have resulted in lab-scale, tabletop lasers which routinely generate pulses few-femtoseconds in duration and multi-terawatt in peak power. This has opened up new domains for scientific exploration by accessing the exotic aspects of light-matter interaction at high intensities. Moreover, in addition to providing rich scientific insights, the interaction of intense laser pulses with solids, liquids and gases have many technological applications as well.

Ultrafast light bursts, comprising a few oscillation cycles of electromagnetic fields, even with relatively low pulse energy, when focused, give rise to unprecedented light intensity levels with extreme temporal resolution. A light pulse is termed ultrashort (or ultrafast) if its temporal duration is less than a picosecond. At moderate intensities ($< 10^{12}$ W/cm$^2$) the electric field of the interacting electromagnetic radiation is much weaker than the static atomic Coulomb fields. In this regime the atomic quantum states get only moderately perturbed for non-resonant excitations, and resonant electron excitation is characterized by bound-bound transitions. Conventional Nonlinear Optics deals with optical processes in this regime. Light intensities larger than $10^{12}$ W/cm$^2$ are considered intense, whose electric field is strong enough to affect most atomic and molecular systems non-perturbatively. At intensities above $10^{14}$ W/cm$^2$ electrons are liberated from atoms and molecules by multi-photon excitations and tunneling through the Coulomb barrier. Higher fields suppress the Coulomb barrier potential further and free the bound electrons to the continuum over the barrier. Under such high irradiation matter will exist in the ionized state, known as plasma.
In this thesis, an experimental investigation of the interaction of moderately intense and intense laser pulses with novel forms of matter, namely metal nanostructures and plasmas, is presented. Initially, the nonlinear optical properties of metal nanostructures are investigated using moderate intensity laser pulses. A new method for estimating the sign of refractive optical nonlinearity in a nonlinear film using a CW laser is then presented. The use of white-light z-scan for measuring the spectral dispersion of optical nonlinearity in a silver nanoparticle solution is discussed afterwards. Later on, the focus of the study shifts to the application of intense laser pulses for creating plasmas in solid and liquid targets, and their spectroscopic study in the x-ray region. For the experiments, we used a He-Ne laser, 7 ns laser pulses from an Nd:YAG laser, and 300 ps, 20 ps and 100 fs laser pulses made available from an ultrafast Ti:Sapphire laser.

A general introduction to the nonlinear optical properties of matter in a strong laser field is presented in chapter 1. Depending on the light intensity the medium shows a nonlinear response that is either moderate or extreme. In this chapter various nonlinear processes such as sum frequency generation, harmonic generation, parametric amplification, optical Kerr effect, optical phase conjugation, stimulated Raman and Brillouin scattering and multiphoton absorption are briefly mentioned. The chapter gives a brief overview of the generation of ultrafast laser pulses. Q-switching and mode-locking methods are discussed. The chirped pulse amplification (CPA) technique used for amplifying ultrafast pulses is explained. Applications of ultrashort laser pulses like ultrafast spectroscopy and TeraHertz frequency generation are mentioned. Furthermore, a general description of intense laser-matter interaction and some of the fascinating applications are presented in this chapter.

A study of the nonlinear optical properties of metal nanoparticles and nanowires is presented in chapter 2. Nanostructures are of interest in optics, and metal nanostructures in particular, because of the surface plasmon resonances they exhibit in the visible region of the electromagnetic spectrum. Various mechanisms controlling the nonlinear light transmission, like saturable absorption, two- and three-photon absorption and reverse saturable absorption are discussed initially. Subsequently, the nonlinear transmission behaviour of Ag@ZrO$_2$-PVA
nanocomposites, and Te and Ag$_2$Te nanowires, measured by the z-scan technique, are discussed in detail. These results show that metal nanostructures possess a large potential for optical limiting applications. In general, the combination of different limiting mechanisms improves the overall limiting performance. The excellent optical limiting properties of nanoparticle-embedded solid-state thin films indicate the possibilities of the design and fabrication of commercially viable optical limiters. Towards the end of the chapter, measurement of the third order nonlinear susceptibility ($\chi^{(3)}$) of silver nanoparticles using the degenerate four-wave mixing (DFWM) experiment in the folded boxcars geometry is discussed. Results show that silver nanoparticles have very good figure of merit (F) values for third order optical nonlinearity.

In chapter 3, a fast and simple method to determine the sign of the nonlinear coefficient $n_2$ in films with high nonlinearity is presented. The technique is similar to the conventional z-scan, except that no aperture is necessary here to determine the sign of the refractive nonlinearity. The method can be considered as a 'visualization' of the z-scan technique for highly nonlinear films. It exploits the effect of spatial self-phase modulation (SPM) and the role of wavefront curvature that leads to a specific far field pattern that is dependent on the sign of $n_2$. When a Gaussian laser beam passes through a thin nonlinear medium like a liquid crystal, the laser beam can induce an unusually large refractive index modulation due to molecular reorientation, and the far field pattern appears as a set of concentric bright and dark diffraction rings. Since the number of SPM principal rings is directly related to the value of the induced birefringence, the method proposed allows a simple evaluation of both the sign (by means of the SPM fine structure) and the magnitude (by counting the SPM principal rings) of $n_2$. The method is independent of the type of nonlinearity that causes the ring pattern, as demonstrated by our measurements in different types of nematic cell, and is expected to work for every kind of highly nonlinear medium.

Chapter 4 discusses a recent application of the femtosecond white-light continuum (WLC) for measuring the spectral dispersion of optical nonlinearity in materials. The dispersion of the two-photon absorption coefficient in the 500-700 nm region in silver nanoparticles is measured using this method. Nonlinear
phenomena like self-phase modulation, stimulated Raman scattering, cross- and induced-phase modulation and self-focusing are responsible for WLC generation in a nonlinear medium. When projected onto a screen, the femtosecond continuum appears to the eye as a white disk, often surrounded by a distinct concentric rainbow-like pattern. White-light continuum finds potential applications in femtosecond time resolved spectroscopy, optical pulse compression, seeding of optical parametric amplifiers, optical coherence tomography, frequency metrology, and linear and nonlinear microscopy. Of the several liquid media we investigated for generating white-light continuum, CCl$_4$ and water are found to be the most efficient. Normalized z-scan curves for a range of wavelengths (500-700 nm) are obtained for silver nanoparticle samples from a single open aperture WLC z-scan. The experimental data obtained is successfully fitted to the standard nonlinear transmission equations, and the dispersion of the two-photon absorption coefficient in the wavelength range studied is determined.

The laser system and the diagnostic equipment used for plasma investigations are discussed in detail in chapter 5. The equipment include the ultrafast CPA laser system (Ti:Sapphire), high vacuum chamber, solid target manipulator, target motion controller, mechanical laser pulse picker, X-ray and γ-ray detectors, and multichannel analyzer. At high laser intensities, air itself will act as a nonlinear medium, and hence a vacuum environment is essential for background free measurements. Our vacuum chamber is provided with a differential pumping arrangement, and can be pumped to a pressure of $10^{-6}$ Torr. The intensity autocorrelation method for pulse width measurement, knife-edge technique and imaging for beam spatial profile determination, calibration of the x-ray and γ-ray detectors, and time-gating of the detectors also are discussed in this chapter.

In chapter 6, the plasma experiments conducted in thin planar liquid jets in ambient conditions are discussed. Intense electromagnetic radiation is known to emanate from laser-produced plasmas. Laser-produced plasma is thus a compact source of pulsed radiation, and can be extended to a wide variety of industrial and scientific applications. In this chapter we discuss the spectroscopic study of x-ray emission from an ultrafast laser-induced plasma, generated in thin planar liquid jets
of approximately 250 µm thickness. Laser pulses of 100 fs duration are focused to the jet to obtain intensity levels close to $10^{16}$ W/cm$^2$. Tunnel ionization is the dominant ionization mechanism at this intensity regime. A general description of various mechanisms leading to plasma production by ultrafast laser pulses is presented. Some important physical parameters of the plasma like Debye length and plasma frequency are explained. Energy transfer mechanisms like inverse Bremsstrahlung, resonance absorption and vacuum heating are discussed. Plasma experiments conducted in thin planar liquid jets of pure de-ionized water, a colloidal solution of silver nanoparticles, and an aqueous solution of silver nitrate are presented. Results show that even in the absence of a vacuum, it is possible to get a substantial amount of soft x-rays from an ultrafast laser-produced plasma. A novel way of enhancing the x-ray emission yield and emission energy range by the incorporation of metal nanoparticles into the liquid used in the jet is presented.

The plasma experiments conducted in Nickel targets under high vacuum conditions ($10^{-6}$ Torr) is given in chapter 7. The target is irradiated at an intensity of $10^{15}$ W/cm$^2$. The x-ray emission in the range of 30 keV to 200 keV is measured using a NaI(Tl) detector. The detector is gated in synchronization with the laser to reduce the cosmic ray background. From the x-ray spectrum obtained, the electron temperature of the nickel plasma is calculated.

Chapter 8 presents the general conclusions drawn from the work done for this thesis, and some future perspectives.