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Matter in Intense Laser Fields

1.1 Nonlinear response of matter in a strong radiation field

Study of phenomena, which occur from the modification of the optical properties of a medium by an optical field, is known as nonlinear optics. They are '*nonlinear*' in the sense that they occur when the response of the material to the applied optical field depends in a nonlinear manner on the strength of the electric field. A laser is a typical source that is strong enough to induce nonlinear phenomena in even weakly nonlinear materials. The electric polarization (dipole moment per unit volume) plays a key role in the description of nonlinear optical phenomena, as the time varying polarization can act as a source of new components of the electromagnetic field. Hence the optical response of a material is generally expressed in terms of the polarization, given as a power series in the applied electric field as,

$$\tilde{P}(t) = \varepsilon_0 \left[\chi^{(1)} \tilde{E}(t) + \chi^{(2)} \tilde{E}^2(t) + \chi^{(3)} \tilde{E}^3(t) + ... \right] \qquad (1.1)$$

where the tilde denote a quantity that rapidly varies with time. $\chi^{(1)}$ is the linear susceptibility of the material, and $\chi^{(2)}$ and $\chi^{(3)}$ represent the second and third order nonlinear susceptibilities of the material respectively. This expression is valid for a lossless and dispersionless medium. Here the fields have been taken as scalar for simplicity [Boyd, R.W.]. It is implicit in the above expression that the atomic polarization follows the changes of the driving electric field instantaneously. This relation between the material response and the applied optical field will break down under conditions like '*resonant excitation*' of the system [Boyd, R.W.], and in the presence of a '*super intense laser field'*, where

the applied laser field becomes comparable to the characteristic atomic field strength.

Depending on the initial and final quantum states of the system, nonlinear optical processes can be categorized as *parametric* and *nonparametric*. In parametric processes such as Sum and Difference Frequency Generation, Second Harmonic Generation, Optical Parametric Oscillation etc. the initial and final quantum mechanical states of the system are identical. Or in other words, the population from the ground state can be removed only for a very shot interval in which the system exists in a *virtual level*, which is not an energy eigenstate of the free system but the combined energy state of one of the energy eigenstates and one or more of photons of the radiation field. According to the uncertainty principle, the population will reside in the virtual level for a time duration of $\hbar/\Delta E$ where ΔE is the difference in energy between the virtual level and the nearby energy eigenstate. In nonparametric processes such as Saturable Absorption, Two-photon Absorption, Stimulated Raman Scattering etc. the population will be transferred from one real level to another real level. These two processes differ in that the parametric processes are described by a real susceptibility whereas the nonparametric processes are described by a complex susceptibility. Photon energy is conserved in a parametric process but in a nonparametric process photon energy need not be conserved as energy exchange with the medium can take place.

Light-matter interaction can be considered by a semi-classical picture (in which the matter is quantized and the field is classical) so long as the number of photons in each mode of the electromagnetic radiation is much larger than one. Since for a typical pulsed laser the number of photons per mode is of the order of 10^{19} [Agostini *et al*. 1988], for interaction involving pulsed lasers a semi-classical treatment is always adequate. Quantized field theory also leads to the same results in the limit of the number of photons per mode becoming large. Quantization of field becomes necessary when the number of photons per mode of electromagnetic radiation is of the order of one. The dependence of the dielectric

constant and magnetic permeability of matter on the field strength inherently suggests nonlinearity in Maxwell's equations. This manifests through the nonlinear polarization that depends on the electric field strength as a power series as given in equation 1.1. This nonlinear response was theoretically investigated by Goeppert-Mayer in 1931 in the earlier years of quantum mechanics [Mainfray *et al.*]. But the intensity of the optical field required to observe the effects of nonlinear polarization was in the order of 1MW/cm^2, which was far unattainable by the classical sources of optical radiation. Therefore experimental observation of nonlinear optical phenomena had to wait till 1961 [Kaiser and Garrett], until the invention of lasers. The charm of these high intensity interactions is that ionization of matter is possible with photons of frequency far below the frequency corresponding to their ionization potential, which is in contradiction to the linear photoelectric effect. Many effects, which were too weak to be detected by conventional sources of light existing in the pre-laser era, became observable by the use of high intense laser pulses [Agostini *et al*. 1988].

1.1.1 Laser intensity regimes

At low intensities, the external field of the laser is much weaker than the static atomic Coulomb field of material systems. Under such a situation, the influence of the laser field is only to slightly perturb the atomic quantum states, when the interaction is non-resonant. For visible and near infrared input laser radiations, an intensity less than 10^{13} W/cm^2 corresponds to the perturbative regime for non-resonant interaction. Phenomena related to the nonlinear susceptibilities $\chi^{(2)}$, $\chi^{(3)}$, $\chi^{(5)}$ etc. take place in this intensity region, where bound-bound transition is the prevailing mechanism. The valence electrons are still bound to the core in these interactions. Since the intensity of the interacting electromagnetic field is moderate and the velocity of the electrons is not in the relativistic regime, the magnetic field component will not give any observable effects. Thus a nonlinear electron oscillator model can represent the atom.

For intensities from 10^{13} W/cm^2 to 10^{14} W/cm^2 *multiphoton ionization* by the simultaneous absorption of many photons becomes significant. This is the intensity regime between the perturbative and strong field regimes of light-matter interaction. In this process, free electrons are generated with a kinetic energy that is a fraction of the photon energy [Shakeshaft, R.]. Any energy in excess of ionization becomes the kinetic energy of the liberated electron. If the ionization energy exactly matches with the energy of an integer number of photons in the electromagnetic radiation, then the resultant electron in the continuum will not possess any kinetic energy. This is the nonlinear counterpart of photoelectric effect in the linear regime of electromagnetic excitation. In this intensity regime the contribution of the freed electrons becomes comparable to that of the induced atomic dipoles pertaining to the bound-bound transitions. A physical example is the self-channeling of intense femtosecond laser pulses over large distances. Here the self-focusing due to optical Kerr effect (contribution from the induced atomic dipole) and self-defocusing due to free electrons tend to balance.

If the strength of the applied laser electric field is comparable to or greater than the binding atomic Coulomb field experienced by the valence electrons, there is a good probability for a valence electron to escape from its bound state via tunneling or above-barrier ionization, before the laser electric field reverses its sign. This electron subsequently wiggles in the linearly polarized laser field and the cycle-averaged kinetic energy of the electron exceeds the binding energy. This regime, in which the ionization process dominates the atomic polarization response, is termed as the *strong-field regime* of nonlinear optics. The nonlinear polarization in this optical ionization regime is mainly governed by the wiggle of the liberated electrons in the proximity of the parent ion. Once the electron is set completely free from the vicinity of the parent ion, it gives only a linear response to the applied laser field yielding results corresponding to classical motion of a free electron in an applied electric field. In the intensity regime of 10^{14} W/cm^2 to 10^{15} W/cm^2 *tunnel ionization* predominates, whereas for intensities greater than 10^{15} W/cm^2 *above-the-barrier ionization* takes over. When the input laser

intensity is extremely high so that the optical ionization leads to the ionization of the inner-shell electrons and gives a higher wiggling energy (comparable to the electron rest mass energy) to the electrons, the interaction regime is termed as *relativistic* nonlinear optics. For the visible and near infrared spectral ranges, intensities of the order and above of 10^{18} W/cm^2 is in the relativistic regime.

1.1.2 Nonlinear effects in the perturbative regime

For laser intensities up to 10^{13} W/cm^2 the response of the medium will be in the perturbative nonlinear regime, provided the applied intensity is higher than that required for a linear response. The nonlinear dependence of the susceptibility on the driving electric field will give rise to phenomena like two photon absorption, three photon absorption, second harmonic generation, sum frequency generation etc [Yariv, A., 1985]. By solving the Maxwell's equations with the time variation of the polarization as the source term, the output from the medium can be found to contain higher harmonics of the fundamental electromagnetic radiation due to nonlinear response of the medium [Milloni & Eberly]. As mentioned before, non-parametric effects like two-photon absorption, three-photon absorption and saturable absorption usually lead to a reduction of energy transmission through the medium at high input laser fields. This nonlinear absorption of laser light can form the basis of novel devices like optical limiters and non-reciprocal light transmitters. In optical limiting, the intensity dependent absorption of laser light is such that at high intensities the material will show a reduced transmission. Optical limiters are passive devices, which have several potential applications. As part of the present work, we investigated nanoparticles of gold and silver for their nonlinear absorption properties. We also studied the nonreciprocal transmission of laser light through materials possessing an axially asymmetric nonlinear absorption coefficient. From the latter studies we found that in general, a saturable absorber and a two-photon absorber (or any material with the property of reduced light transmission at higher laser intensities) placed in

tandem will transmit light in one direction and prevent transmission in the opposite direction, thereby providing an optical diode action.

1.1.3 Optical field ionization of atoms

Conventionally, optics deals with phenomena in the eV regime, where photon energies fall approximately between 1.77eV (700nm) and 3.11eV (400nm). The process by which an electron in the outermost shell gets excited to a continuum is named as ionization. In the linear optical regime ionization happens only when the energy of the incident photon matches with the binding energy of the electron in its shell. Hence electromagnetic radiation with higher photon energy ionizes matter with greater probability. As we move towards the high frequency side of the electromagnetic spectrum, the ionization probability increases with γ-rays as the highest ionizing radiation. Here it is the energy of the individual photon that determines the ionization of a single atom, rather than the intensity of the incident radiation (intensity helps only in ionizing a larger number of atoms). But with the advent of high power lasers, the electric field strength associated with the emission is so high that the radiation intensity has a deciding role in the ionization of a single atom. Thus ionization of atoms or molecules with non-resonant light frequencies becomes possible. The intensity of a light field can be related to its electric field strength through

$$I = \frac{1}{2}\sqrt{\frac{\varepsilon_0}{\mu_0}}E_0^2 = \frac{1}{2}\varepsilon_0 c E_0^2 \tag{1.2}$$

where c (the velocity of electromagnetic radiation in free space) is given by

$$c = \frac{1}{\sqrt{\varepsilon_0 \mu_0}} \tag{1.3}$$

The behaviour of an atom in high laser intensities (>10^{13} W/cm^2) is usually analysed on the basis of the 'keldysh parameter' given as:

$$\gamma = \frac{\omega_0 \sqrt{2mI_0}}{eE_a} \tag{1.4}$$

where ω_0 is the laser carrier frequency, m the electron rest mass, I_0 the ionization potential of the atom (i.e. the binding energy of the most weakly bound electron), E_a the time dependent amplitude of the linearly polarized laser field, and e the electron charge [Keldysh, 1965], [Brabec *et al.*], [Protopapas *et al.*]. For high frequencies and not very high intensities $\gamma>1$ and multiphoton ionization (MPI) dominates. For very high intensities ($>10^{14}$ W/cm^2) and low frequencies $\gamma<1$, and the laser field significantly modifies the Coulomb potential. For the range of laser intensities 10^{13} W/cm^2 to 10^{17} W/cm^2 that are readily available from the present day high power lasers, the corresponding electric field strength is in the range of 10^{10} to 10^{12} V/m [Batani *et al.*]. Here the electric field strength is comparable to the Coulomb field that binds electrons in the atoms or in the valence shell for the molecular bonding. The ionization of matter is a result of different mechanisms in different intensity regimes as discussed in section 1.1.1.

As an extension of the linear photoelectric effect, the nonlinear photoelectric effect, i.e., the multiphoton ionization should absorb N number of photons sufficient to ionize the matter, and the kinetic energy of the ejected electron should possess a kinetic energy not more than the energy of one photon of the exciting laser field. But it is being observed that often the kinetic energy of the ejected electron far exceeds the energy of a photon of the exciting laser-field, which calls for a modified explanation of the multiphoton ionization rather than a direct extension of the linear photoelectric effect to a nonlinear photoelectric effect. This excess absorption of photons can be explained by inverse Bremsstrahlung absorption (if the matter is dense enough to cause sufficient collision) leading to the absorption of photons from the laser field by the accelerating electrons during their scattering with neutral atoms or ions. This phenomenon was observed in 1977 [Weingartshofer *et al.* 1977] in which the electrons are produced by standard multiphoton ionization but the excess of energy is attained by inverse Bremsstrahlung absorption. Multiphoton absorption

and the successive Inverse Bremsstrahlung absorption cannot be applied to the case of low density matter - short laser pulse interaction, as the electron-ion collision probability is very less in such a system [Federov, M.V.], [Agostini *et al*. 1988], [Faisal, 1987]. Under this condition a mechanism in which the electron absorbs more number of photons than that is sufficient for multiphoton ionization, is proposed. This is called *Above Threshold Ionization* (ATI), which was first discovered in 1979 [Agostini *et. al.* 1979]. In the case of low-density matter each electron in the continuum interacts only with its own parent ion or atom and does not undergo any collision with the neighbouring atom or ion. In ATI, when an electron gains energy equal to the ionization potential of the atom, the electron is moved to a distance where the coulomb potential of the nucleus is not significant, and hence it behaves as a free electron. This electron absorbs more photon from the laser field while it is still in the vicinity of the parent nucleus. Therefore the absorption of larger number of photons by low-density materials for multiphoton ionization in an intense field is being described by ATI [Kyrala, G.A.]. For experiments at moderately high intensity, where the bound-state energies of the atom remain un-shifted by the electromagnetic radiation, the perturbative theory of ATI holds [Fabre *et al*. 1983]. The perturbation theory of ATI breaks down at higher laser intensities and for longer wavelengths [Lompre et al., 1985], [Deng and Eberly], [Reiss, H. R.]. The photoemissions under this condition are analyzed using the non-perturbative approach [Krause *et al*.], [Chu *et al.*], [Cormier *et al.*].

For higher intensities of the excitation laser field corresponding to a regime of $\gamma<1$, the Coulomb barrier becomes narrow allowing *Tunnel Ionization* (TI). The γ value decides whether the ionization process is MPI or TI. At still higher intensities ($>10^{15}$ W/cm^2) the electric field amplitude of the laser is so prominent that it suppresses the Coulomb barrier below the energy level of the ground state leading to *Above the Barrier Ionization* (ABI). The minimum value of the exciting laser field that is required to induce ABI can be obtained by equating the maximum value of the resultant atomic potential, in the presence of

the laser field, to the binding energy of the electron. Different routes of ionization of an atom in the presence of an intense laser field are shown in figure 1.1.

The electron, once freed from the Coulomb potential by any one of the above three mechanisms, is released into a strong electromagnetic field provided by the focused excitation laser. Light being a combination of transverse electric and magnetic fields, the released electron sees the two fields and is subjected to the Lorentz force. Since the force exerted by the magnetic field is v/c times less than that of the electric field in the non-relativistic regime, the electron follows the electric field of the laser pulse. Hence the electron will undergo an oscillatory motion in the laser electric field. The oscillatory energy possessed by the free electron in the laser field is known as the *electron quiver energy*.

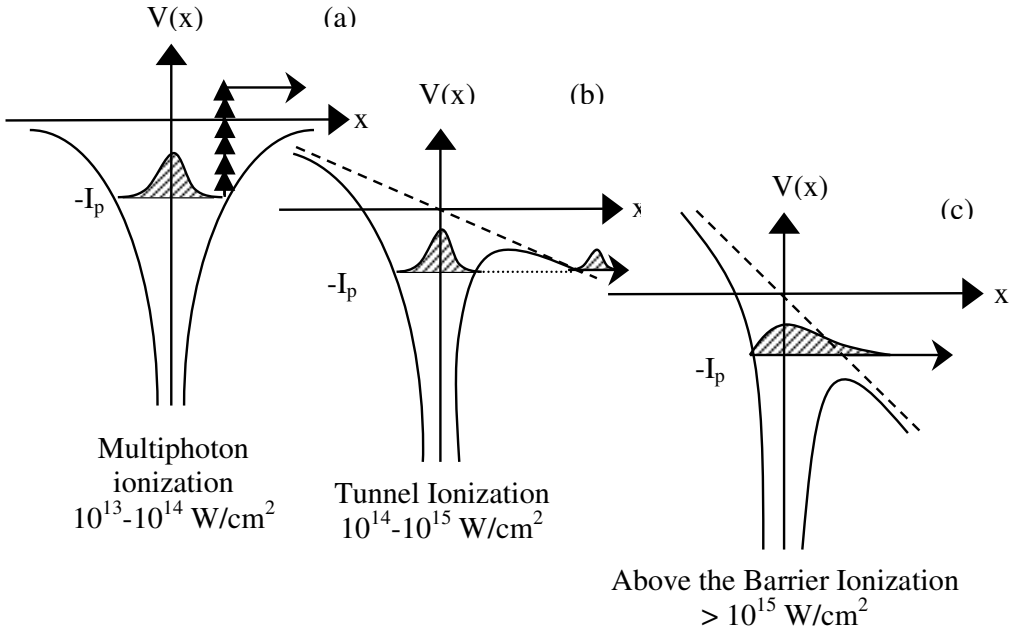

Figure 1.1: Schematic diagram of the three possible ionization mechanisms in the high intensity laser field – atom interaction.

In the nonrelativistic regime, the electron follows the laser electric field in direction and frequency. In the relativistic regime, the magnetic field will also

start to exert a significant force on the electron so that the resultant trajectory of the electron in the laser field will be governed by both the electric and the magnetic fields [Maltsev *et al.*]. The quiver energy of the electron in the laser electric field is given as

$$E_{osc} = \frac{e^2 E^2}{4m\omega^2} \tag{1.5}$$

where E is the local instantaneous electric field and ω is the angular frequency of the laser field.

Apart from the electron quiver energy due to the oscillation of the electron in the laser field, there is another energy that is acquired by the free electron subjected to an inhomogeneous laser field. This is named as *ponderomotive energy*. The concept of ponderomotive force is well known in the electrodynamics of condensed media [Landau & Lifshitz] arising from the inhomogeneity of the medium or that of the field. For a less dense medium the origin is the inhomogeneity of the field itself. In the case of a pulsed Gaussian laser beam, there is an inhomogeneity in the laser field in space and time. The electron motion in an inhomogeneous electric field when averaged over fast oscillations in the field gives the averaged trajectory of the electron in the inhomogeneous field, and is interpreted as the motion under the ponderomotive force [Federov], [Zhu *et al.*]. This is the way in which the electron decouples from the laser focal volume [Bucksbaum *et al.*]. The ponderomotive force, which pushes the electrons away from the region of high field pressure, is given as

$$\vec{F}_p = -\frac{e^2}{4m\omega^2}\vec{\nabla}\vec{E}^2(x) = -\vec{\nabla}U_p \tag{1.6}$$

for a linearly polarized laser field oscillating with an angular frequency ω. U_p, the ponderomotive energy is the cycle averaged quiver energy of the electron in the oscillating laser electric field [Gaponov & Miller], [Kruer W.L.].

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Even though the above discussion of optical ionization processes mainly pertain to diffuse atomic systems, the ionization mechanisms remain the same for molecular states too. The only difference is the relevant change in the energy

levels and ionization potentials of the system. One qualitative difference is that, in intense field molecular ionization, the ionization rate sensitively depends on the molecular configuration, i.e., inter nuclear distance. Hence the ionization can act as a sensitive probe for the molecular structure and motion. [Eberly et al., 1978], [Seidemann et al., 1995], [Chelkowski et al., 1996].

1.2 Generation of intense ultra short laser pulses

The first laser, invented by Maiman in 1960, based on a ruby crystal and pumped by a xenon flash lamp, had an output lasting between a microsecond and a millisecond [Bloembergen, N.]. Ever since, the reduction of laser pulse width has become a frontier field of research in laser engineering. The shortness of the pulse duration opened a new vista of temporal regimes to be explored. Time resolved studies have become mainstream in the investigation of different systems in chemistry, biology and physics. The confinement of energy into very short time and space leads to yet another regime of studies where 'tiny stars' have been created. The radiative energy density, pressure, temperature, electric and magnetic fields become comparable to those observed in the thermodynamically driven stellar interiors. Optics, which had faced somewhat of a saturated state in the beginning of the 20^{th} century, got a new birth due to the invention of lasers and the emerging technologies that squeezed laser pulses to shorter regimes in space and time. Part of the work reported in this thesis could be done only because of the availability of very short and intense laser pulses. It will be worthwhile to sketch a short outline of the generation of intense short laser pulses in this context.

The production of laser pulses existing for a few nanoseconds was first suggested and demonstrated by Hellwarth in 1961. The switching of the quality factor of a cavity from a low Q value to a high Q value (high loss to low loss) routinely produces laser pulses of a few nanoseconds pulse duration [Siegman, A. E.].

Most of the intense pulses that belong to the ultrashort pulse category are those with pulse durations in the picoseconds to femtoseconds regime. The principle behind the generation of these pulses is called '*mode locking,*' [DiDomenico], [Hargrove *et al.*], [Yariv, A.] which is a 'temporal' interference phenomenon. Here the simultaneous oscillation of a very large number of coherent, phase-locked longitudinal modes (the different frequencies existing inside a cavity) in a laser gives a resultant field that is nearly zero most of the time, except for a very short interval of time. As a result of the short lived temporal constructive interference between the different longitudinal modes, the entire energy is concentrated to these very short periods [Brabec & Krausz]. The electric field distribution in the laser cavity with N longitudinal modes in phase with the initial phase chosen to be zero is given as:

$$E(t) = \sum_{n=0}^{N-1} E_n e^{i(\omega_0 + n\Delta\omega)t} \propto \frac{e^{iN\Delta\omega t} - 1}{e^{i\Delta\omega t} - 1} e^{i\omega_0 t} = \frac{\sin(N\Delta\omega t/2)}{\sin(\Delta\omega t/2)} e^{i\omega_0 t} \qquad (1.7)$$

In a laser cavity the longitudinal modes are equally spaced with the frequency spacing $\Delta\omega$ given by $c/2L$, where L is the cavity length. ω_0 is the central frequency [Rulliere, C.], [Diels & Rudolph]. In order to get a short pulse a very large number of the cavity longitudinal modes should be locked together. The shortest pulse that can be achieved from a lasing medium of linewidth $\Delta\nu_g$, is $1/\Delta\nu_g$. The intensity of the mode locked output from the laser is given by

$$I(t) \propto |E(t)|^2 = \frac{\sin^2(N\Delta\omega t/2)}{\sin^2(\Delta\omega t/2)} \qquad (1.8)$$

In most of the high power, short pulse lasers, pulse generation and amplification are done in steps. The generation of the femtosecond pulse follows the principle of mode locking as described above with various active and passive modes of realization, of which the details can be found elsewhere [Brabec & Krauz], [Millonni & Eberly]. In the 1970s and 80s successful passive mode locking was achieved in dye lasers [Fork *et al.*], [Valdmanis & Fork], [Shank & Ippen]. Ti:Sapphire (Ti:Al_2O_3), the solid state laser material having a broad gain bandwidth to support femtosecond pulses, was discovered in the late 80s

[Moulton]. Passive modelocking in this solid-state laser was observed to exist without the necessity of an intracavity saturable absorber [Spence *et al.*], and was termed as 'magic modelocking' in the initial days. This was later theoretically understood as Kerr Lens Modelocking (KLM), which is the major mechanism in the present-day solid-state femtosecond lasers for producing ultrashort pulses [Keller *et al.,* 1991], [Salin *et al.*] ,[Curley *et al.*]. This is based on the occurrence of self focusing by the intensity dependent index of refraction (the Kerr effect) in the Ti:Sapphire crystal. In combination with a suitable aperture, the round trip gain is increased at higher intensities as more light is focused to pass through the aperture. A Schematic representation of KLM is given elsewhere [Brabec & Krausz], [Keller, 2003]. Since the modelocked femtosecond pulse needs to have many frequencies locked together, group velocity compensation is very important in generating very short pulses, especially sub 10fs pulses [Zhou *et al.*], [Jung *et al.*], [Xu, L. *et al.*], [Sutter *et al.*], [Ell *et al.*], [Steinmeyer *et al.*].

The energy per pulse of the mode-locked pulse train in the oscillator will be usually of the order of a few nanojoules. In order to increase the energy, the femtosecond pulse should be amplified. For this, small-scale high power lasers [Perry & Mourou 1994] basically utilize a technique called 'Chirped Pulse Amplification (CPA)'[(first demonstrated for microwave) Cook,C.E.,1960], [Strickland & Mourou, 1985 (first optical CPA demonstration)], [Maine *et al.*, 1988]. In this technique, the low energy femtosecond laser pulse from the oscillator is initially stretched in time [Pessot *et. al.*, 1987], [Champaret *et al.*], which is then amplified. The stretching is done to avoid nonlinear effects like self-focusing in the solid-state amplifiers, which will otherwise damage the amplifying medium. The stretcher introduces a frequency dependent phase to all frequency components of the femtosecond pulse from the oscillator. Usually a combination of a grating and a mirror is used to stretch the pulse. The grating directs different frequencies at different angles, and the inclination between the grating and the mirror is such that the fastest frequency has to travel longer distance and the slowest frequency has to travel shorter distance, so that the pulse will stretch in

time due to positive dispersion. Thus the pulse is positively '*chirped*', that is, it has a time dependent instantaneous frequency, where the slow frequencies appear first and the fast frequencies follow. The frequency content of the pulse remains the same as that from the oscillator. In the 'chirped pulse' the frequencies are only temporally separated. The optics in the amplifier through which the pulse passes will give an additional positive dispersion. Thus the initial positive dispersion keeps the pulse stretched throughout the process of amplification, thereby avoiding the possibility of damage of the gain medium and other optical components. The stretched pulse is usually amplified by passing it through a regenerative cavity and a multi pass cavity, both of which have Ti: Sapphire as the gain medium. The pulse is compressed right after amplification by introducing negative dispersion using a grating compressor, where the fast frequencies are given shorter paths and slow frequencies are given longer paths. After the introduction of CPA, a large variety of solid state materials were tried as active media for the generation of intense ultrashort pulses [Perry *et al.* 1990], [Pessot *et al.* 1989], [Kmetec *et al.*], [Patterson *et al.*].

The very high intensity attainable by these ultrashort laser pulses leads to unprecedented acceleration of electrons in a very short length compared to the long path acceleration in accelerators, leading to the realization of 'table top' electron accelerators [Tajima *et al.*] [Umstadter *et al.*], [Siders *et al.*]. An intense laser field interacting with a solid target produces high-energy proton beams [Clark *et al.*] making it a possible compact proton source. One of its application can be to produce radioactive sources used in Positron Emission Tomography (PET) in nuclear medicine. Detailed description of the application of ultrashort lasers for different nuclear phenomena can be found in [Lendingham *et al.*]. With the availability of high intensities from pulsed lasers, different types of interactions can be studied. A schematic of the different processes at different laser intensities is shown in figure 1.2.

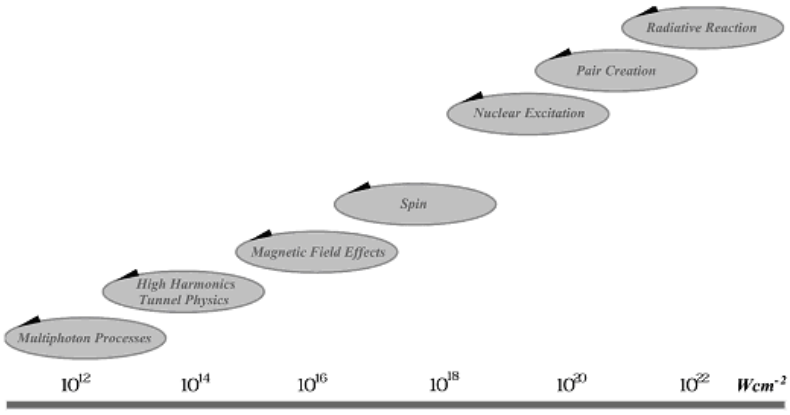

Figure 1.2: Schematic of different processes associated with the interaction of intense laser pulses with atoms [Keitel].

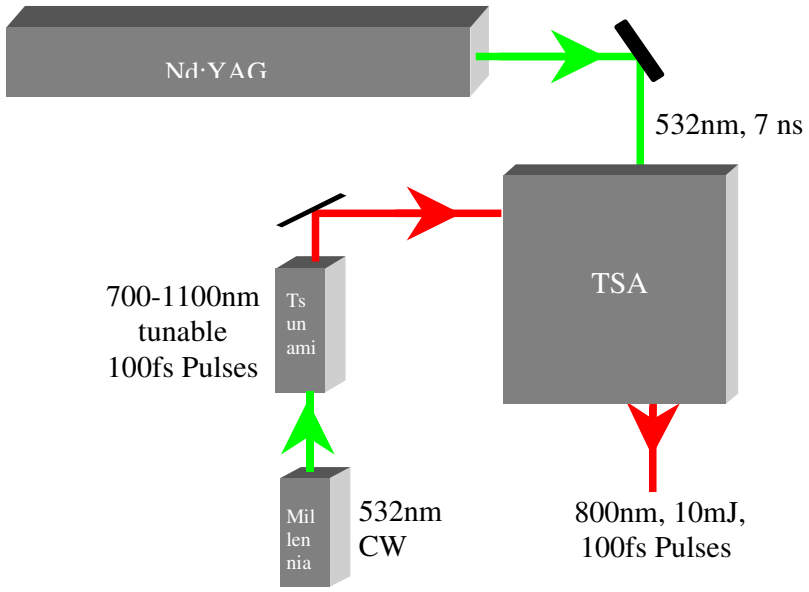

Figure 1.3: Schematic of the femtosecond laser system [Spectra Physics] used for the studies reported in this thesis.

1.3 Outline of the thesis

In this thesis we have investigated the interaction of moderately and highly intense laser pulses with metal nanoparticles and plasma respectively. An

introduction to the nonlinear response of matter to large electric fields present in intense laser pulses is outlined in the first part of this chapter, followed by a discussion of the different mechanisms of laser-matter interaction in different regimes of laser intensity. The nonlinear effects in the moderate intensity regime are discussed in brief. The optical field ionization mechanisms are outlined. The methods for generation of nanosecond and femtosecond laser pulses are described. Chapters 2 and 3 contain results from the moderate intensity laser field-matter interaction studies. Chapter 2 contains nonlinear light transmission studies in metal nanoparticles of different sizes, dispersed in different host materials. Experiments as well as numerical analysis are presented. The materials studied are found to be potential candidates for application as fast passive optical limiters to protect eyes and sensitive sensors from optical damage. Chapter 3 discusses a new idea for the design and implementation of a passive all-optical diode. Here, nonlinear absorbers are used for realizing the optical diode action. Both experimental results and numerical simulation results are presented. In chapters 4 and 5 the creation of plasma using femtosecond laser pulses of intensities in the order of 10^{15} W/cm^2, and the interaction of the coherent laser pulse with the generated plasma are studied. Chapter 4 contains a spectroscopic analysis of the emission resulting from the interaction of 100fs laser pulses with a planar water jet of 250μm thickness at normal incidence. Chapter 5 discusses the emission of second harmonic radiation of the exciting laser field, and incoherent emission in the visible, from plasma produced in the planar water jet for oblique incidence of the exciting laser pulse. Chapter 6 describes the design and fabrication details of an experimental setup to study the interaction of ultrashort laser pulses with liquid droplets. This includes the design details of a differentially pumped vacuum system that handles injected liquid load, the calibration of solid state x-ray and γ-ray detectors, synchronization of the scintillation detector with the laser, and imaging of the liquid droplets in vacuum. The conclusions drawn from the experiments done and future prospects are given towards the end of the thesis.