Preface

Studies on multiphoton process has gained momentum after the advent of lasers in 1960, but the history traces back to 1930's when Göppert-Mayer developed the theory on spontaneous and stimulated two-photon process based on second order perturbation theory. It is well known that even 'Rayleigh and Raman scattering' are bona fide two-photon process. Invention of laser led to the observation of various higher order process like stimulated Raman scattering, higher harmonic generation, multiphoton ionization (MPI) and selective excitation of atoms, which became topics of immense interest to both theoreticians as well as experimentalists. Unlike the single-photon transitions, multiphoton processes depends on intensity and polarization of the incident radiation. This intensity dependence makes this process a nonlinear one. Area of multiphoton ionization is rich with many interesting phenomena. Multiple ionization of atomic system was observed in 1975 and then follows the experimental observations of electrons with large kinetic energies which can not be accounted by the single photon absorption. The area became more interesting with the experimental observation of ac-Stark shifts, tunneling ionization and recently the atomic stabilization in the presence of intense laser fields.

The availability of lasers with intense-short pulses, leads to the possibility of using multiphoton spectroscopy for probing the fundamental physics
of atom-field interaction. The availability of increasingly intense lasers has also made possible the observation of a wide variety of multiphoton processes, including multiphoton ionization, harmonic generation and laser-assisted electron atom scattering. Since the laser pulses are so short the electric field can oscillate only a few times during the pulse width time interval, the tunnelled electron wave packet moves under the influence of the strong field, when the electric field switches it can re-collide with its parent ion. This electron can interfere or diffract with the bound state wave function and can even scatter from its parent ion elastically or inelastically and excite it.

Multiphoton process corresponds to higher order terms in the perturbation theory of matter-field interaction. Perturbation theory proves to be valid for intensity of light up to $I = 10^{13}$ Wcm$^{-2}$. It was found that at moderate intensities, direct multiphoton ionization as well as above threshold ionization (ATI) are satisfactorily explained by perturbation theory. The process of ATI is the absorption of more number of photon than the minimum number required for the ionization, thus leading to transitions among the continuum states. ATI can be studied using high resolution photo-electron spectroscopy.

In the present thesis, we concentrate on various multiphoton process in hydrogen atom. This study on hydrogen atom provide, a basis for the studies on a large number of atomic systems like alkali atoms, negative ions etc. We restrict our work on multiphoton process which can be treated in the framework of perturbation theory. Under the framework of perturbation theory, we also studied above threshold ionization (ATI) for the case of two- and three-photon ionization.

The introductory chapter (Chapter 1) is a review on various multiphoton processes, starting from the description of simple one-photon ionization. We reviewed the main topics in atom-field interaction covering both theoretical and experimental investigation of the subject over a broad range.
We tried our best to discuss various experimental observations and its theoretical explanations, making the review a complete one.

This includes the effect of intensity on ionization processes, polarization dependence of MPI, angular distribution of the ejected electron, multiple ionization and resonance enhanced MPI. A separate section is added for the discussion on tunneling ionization, which is entirely independent of multiphoton ionization physics. We dedicate separate sections for describing the observed nature of ionization spectrum in different atomic systems other than hydrogen atom, which includes theoretical models for describing alkali atoms and atoms with many valence electrons. We conclude the chapter by discussing the recent developments in the superintense laser physics. In each sections we tried to describe the experimental observation using the available theoretical explanation on the subject.

In Chapter 2 we mainly concentrate on the theoretical methods available for the description of multiphoton ionization. The range of intensity considered is such that the perturbative treatment is feasible. In the first part we highlight the problems arising in the formulation of multiphoton ionization using perturbation theory, especially problem of infinite summation appearing in the perturbative expansion. These infinite sum of intermediate states includes discrete as well as continuum states. Then few methods are described where the infinite sum is approximated using various methods and we also reviewed two methods which avoid the explicit summing of the contribution from intermediate states.

First the expression for two-photon transition probability is derived using perturbation theory in the dipole approximation. Then the available methods are reviewed. The approximate methods used in perturbation theory are technique by Bebb and Gold, truncated summation, variational treatment, Green’s function method and Dalagarno-Lewis method. Non-perturbative treatment includes WKB method and R-Matrix Floquet method. Among these Green's function method is extensively used for per-
turbative and for high intense fields R-Matrix Floquet method is widely used.

Chapter 3 consists of mainly a discussion on second order processes, where we have grouped together a few processes which can be explained using second order perturbation theory. Starting from the second order matrix element we illustrated the application of the Dalgarno-Lewis procedure. The analytical expressions obtained can be used for various two-photon processes involving the hydrogen atom.

Various two-photon processes includes bound-bound transitions, two-photon ionization, ac-Stark effect and elastic scattering of photon from the hydrogen atom. In the section where two-photon bound bound transition is discussed, we have calculated the transition probability for a two-photon transition to occur from the ground state to any excited state, with the quantum numbers $n$ and $l$. The section for two-photon ionization deals with the evaluation of the scattering cross section for two-photon processes. This includes both 'direct' and 'above threshold ionization'. We have formulated the method, derived the relevant amplitude expressions and also described the origin of two-photon selection rules. We have extended the range of applicability of the relevant expression for all values of incident photon energy, by the process of analytical continuation.

In presence of intense laser fields, the atomic levels are shifted or broadened and these stimulated radiative corrections are known as the ac-Stark shift. This is one of the interesting subject in the intense laser field science. We have done a perturbative treatment for the ac-Stark effect experienced by a non relativistic hydrogen atom irradiated by a mono mode laser field. Here we obtained an alternate expression for dipolar polarizability and it was found that at the limit where the frequency of the incident photon goes to zero corresponds to the static polarizability of the hydrogen atom. Then the dispersion relation of Kramers and Heisenberg is evaluated in our formalism. Since both are two-photon processes, we used a unified treatment
to obtain the alternate analytical expressions and the results are compared with values reported in the literature.

In **Chapter 4** we discuss three-photon processes. The first one which we discuss is the three-photon transition probability from ground state of atomic hydrogen to any excited bound state allowed by three-photon selection rules. Secondly we obtained scattering cross section for three-photon ionization of atomic hydrogen. Three-photon ionization includes three separate cases depending on the energy of the incident photon. They are direct ionization, above one-photon and two-photon ionization threshold. Finally we have discussed analytic continuation, which is mainly used to describe above threshold ionizations.

From the analytical expressions for three-photon radiative transition matrix elements the cross section for three-photon ionization of atomic hydrogen is calculated for both linearly and circularly polarized light with a wide range of photon energy spectrum including the near resonance and numerical comparison is made with the values obtained by different methods. We dedicate a separate section for the description of above threshold calculation which is obtained by the analytical continuation of the relevant expressions previously obtained. Taking the expression for two-photon processes we also demonstrate the process of analytical continuation in detail.

**Chapter 5** contains concluding remarks which summarize our work. The future prospectives are also discussed.