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Interaction of atoms with intense laser fields†

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In recent years, new, sometimes counterintuitive, phenomena have been discovered in atom-laser interaction, which covers from multiphoton ionization and high harmonic generation in an intense laser field to the suppression of ionization in a superintense field. Traditional perturbative approaches cannot be resorted to for explaining these phenomena. In this article, various non-perturbative approaches developed over the years are discussed. Experimental aspects relating to these phenomena are also described.

Contents

1. Introduction

Einstein's photoelectric law adequately describes the ionization of matter but is valid only for the incident light beam having a low density of photons. With the advent of lasers, a source with a density of millions of photons, multiphoton processes came into prominence and ionization processes became number-dependent, i.e. dependent on the laser intensity rather than the frequency[1–7]. A typical laser of intensity 10^{14} W cm^{-2} (1 a.u. of intensity = 3.5×10^{16} W cm^{-2}), having a wavelength of 1064 nm, carries a photon density of 2×10^{10} photons per cubic wavelength. Under such a high intensity, an electron in an atom or a molecule absorbs additional photons than were required for ionization and the emission process involves the ejection of photons having energy in odd multiples of the incident laser frequency (for centrosymmetric systems under a dipole approximation).

These new unexpected phenomena resulting from multiphoton processes cannot be adequately explained by perturbative approaches as the states are highly 'dressed' by the strong laser field. Over the last two decades, several non-perturbative methodologies, both time-independent and time-dependent, have been developed which are both physically insightful and computationally viable.

In this article, we review the basic principles involved in various multiphoton processes. Section 2 describes the experimental aspects of the processes concerned while a brief description of the theoretical treatments is presented in Section 3. Finally, Section 4 speculates on the likely future challenges in this rapidly widening frontier of physics and chemistry.

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2. Experimental observations

2.1. Multiphoton ionization (MPI) :

MPI of free atoms is primarily a transition from a bound state to a continuum, with incident photon energies much smaller than the ionization potential. Since this usually occurs at intensities $>10^{13}$ W cm^{-2}, the perturbation of atomic states, known as AC-Stark shifts, are large and must be taken into account. The 'non-perturbative' aspect of AC-Stark shifts conflicts with perturbation theory[4] which describes N-photon ionization by a perturbation rate P_N given by

$$P_N = C_N I^N \qquad (1)$$

where C_N is the generalized cross-section.

A free electron of mass m_e and charge e in a laser electric field of peak amplitude E_0 possesses a kinetic energy due to its oscillation (quiver) in the laser field. This cycle-averaged kinetic energy is known as ponderomotive energy (quiver energy or wiggle energy) and is given by

$$U_p = e^2 E_0^2/(4 m_e \omega_L^2) \qquad (2)$$

where ω_L is the incident laser frequency.

The oscillation of a free electron due to oscillating fields is complicated by the fact that since a laser beam brings in magnetic as well electric fields, the electron's trajectory is not simply given by one-dimensional harmonic motion in response to the electric driving force. Furthermore, the spatial and temporal (time – varying) inhomogeneities in the applied fields give rise to large effects observed in high intensity laser-electron interaction. MPI experiments in high-intensity are divided into two classes–those which are sensitive to the ponderomotive forces and those which are not.

For any multiphoton ionization process, there exists a saturation intensity which is defined as the intensity under which the corresponding rate times the pulse duration is close to unity, i.e. for a pulse of given duration, there is a highest intensity above which no more ionization can occur since all the sample atoms have been ionized. In other words, no atomic system can survive an exposure to such an intensity for times much longer than the pulse duration. To study the behavior of atoms under a given intensity, the saturation intensity must be increased at least to this value, by correspondingly reducing the pulse duration. During the last decade, new laser technologies emerged for the production of femtosecond (1 a.u. of time = 0.0242 fs, 1 fs = 10^{-15} s) and even subfemtosecond pulses[8,9]. Short pulses acquired an even greater importance when it was realized that they have another extremely useful property, namely, that they suppress all the ponderomotive effects, which can sometimes obscure the analysis of MPI electron energy spectra.

2.1.1. Above threshold ionization (ATI) : When ionization occurs via more than the minimum number of photons required (threshold), it is called ATI. A well-resolved ATI spectrum shows several peaks at lower intensities separated by the photon energy, with the high energy peaks having intensity several times less than lower energy ones. As the intensity increases, the lowest order peak is reduced in magnitude and vanishes for the highest intensity (Fig. 1). At intensities exceeding 10^{13} W cm^{-2} for infrared lasers, MPI always yields an ATI-like spectrum.

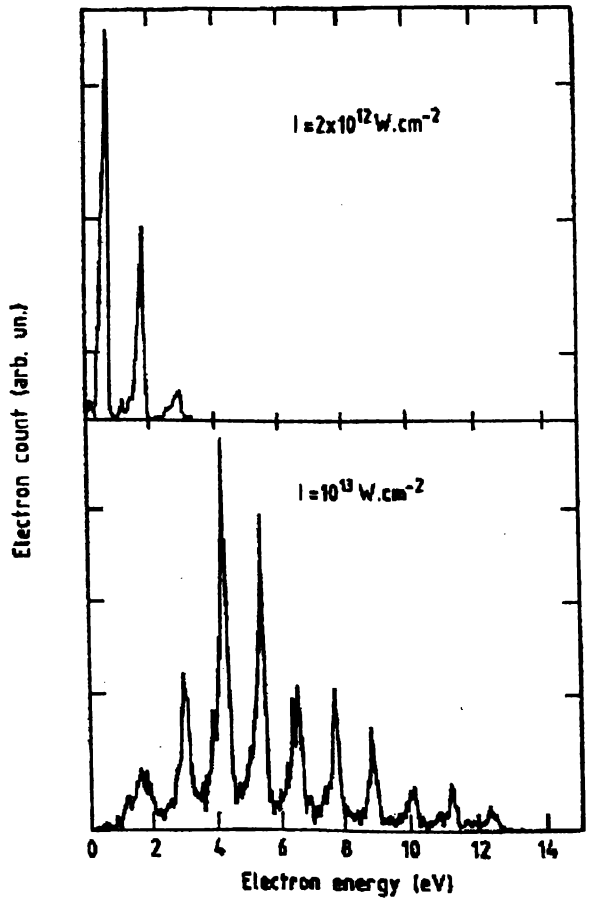

Fig. 1. Experimental ATI spectra of xenon at 1064 nm for laser intensities : (a) $I = 2 \times 10^{12}$ and (b) $I = 10^{13}$ W cm^{-2}. The electron count is plotted as a function of electron energy (in eV). The peaks are separated by $\hbar\omega_L$. (Reproduced by permission from H. G. Muller, P. Agostini and G. Petite, p. 12 in Ref. 1).

The ponderomotive effects are responsible for the 'peak suppression' in ATI spectra. This is a non-perturbative effect. Since the total energy of a free electron oscillating in a laser field is $U_p + E_T$, where E_T is the translational energy, the continuum states shift upwards relative to the lower bound states, approximately by U_p and produce an increase in the ionization threshold. This AC-Stark shift increases with intensity resulting in peak suppression.

The fact that the ionization limit, the Rydberg states (higher energy states near continuum) and continuum states shift in this manner is not a property of intense light. As the ionization limit is increased in a laser field, more photons must be absorbed with the excess energy going into the oscillation of the photoelectron. The oscillatory ponderomotive energy is converted to translational kinetic energy as the electron leaves the focus of the laser beam. Thus, the ponderomotive energy does not alter the energies of the detected electrons and no electron can be observed with an energy less than the U_p. If the laser pulse is so short that the electrons do not leave the focus while the light is present, different results are obtained.

For ATI experiments, mainly rare-gas atoms have been placed under the focus of a laser beam. These atoms have deep ground states with the lowest lying excited states separated by many quanta of photon energy. Therefore, the quiver energy of their ground state is negligible, while that of the Rydberg and continuum states is free-electron-like. This results in an approximate increase of ionization threshold by U_p.

A photoelectron spectrum records the actual photoelectron energies only in the limit of very short pulse duration. The major results of ultra-short pulse lasers, in the study of high intensity MPI, is the removal of masking effects of ponderomotive forces on the free electrons leaving the focus of the laser beam.

2.1.2. Ionization rates : When the electronic quiver amplitudes are large compared to the size of an atom, the probability for an electron to find its way back to the atom after being ejected, becomes negligible, thereby limiting the total ionization rate. When the two are comparable in size, this probability may be appreciable. The absorption of excess photons does not manifest itself in any way on the total ionization rate.

Even a laser field of moderate intensity can cause a large quiver amplitude when the frequency is low (eq. 2), making ionization stage-independent from the above-threshold stage, the former determining the energy distribution of the ejected electrons. The absorption of excess photons is associated mainly with the acceleration of the free electrons by the laser field. A free electron will always acquire the quiver energy without difficulty, no matter how many excess photons are absorbed. Therefore, the absorption of excess photons occurs spontaneously to overcome the corresponding increase in ionization potential (whatever the quiver energy may be) and has no effect on the rate of the overall process.

2.1.3. The way the electron becomes free : The process of making an electron free depends on the experimental situation in which MPI is observed. There are two distinct ways to bring about ionization, the first involving only a few photons and the second involving a high density of photons. Through the first, called 'multiphoton absorption', the absorption of a few photons leads to the production of electrons with kinetic energy close to the binding energy in the atom. At intensities $<10^{14}$ W cm^{-2}, this is the most preferred way for multiphoton excitations.

An alternative way opens up[10,11] when a strong enough low-frequency incident field distorts the atomic potential to such an extent that a potential barrier is formed through which the electron can tunnel. This tunnelling description is more suited when a large number of photons are involved. In this case, the electrons emerge with positive kinetic energy only after tunnelling through the potential barrier. As the field strength is increased, the potential gradient becomes increasingly more negative, the barrier becomes progressively lower and finally a situation arises where the ground state is no longer bound. This is popularly known as 'over-the-barrier' (OTB) ionization and occurs at $\sim 10^{14}$ W cm^{-2} for a low-frequency laser. The newly created free electron acts as a classical point charge in a laser field with a certain initial velocity and direction determined by the phase of the field. Hence, very rapid ionization takes place near the peak of the pulse, where the barrier is most suppressed. This tunnelling picture has been verified by a number of experiments. Keldysh[10] had defined a parameter Γ which is an indicator of the way the electrons become free,

$$\Gamma = [U_I/2\ U_p]^{1/2} \qquad (3)$$

where U_I is the ionization potential and U_p is given by eq. (2).

If $\Gamma < 1 \rightarrow$ Tunnelling ionization

If $\Gamma < 1 \rightarrow$ Multiphoton absorption

i.e. multiphoton dynamics dominate when $\Gamma > 1$, otherwise the electron would choose another way. An ATI spectrum reveals these features clearly. The spectrum becomes noisy as $\Gamma \rightarrow 1$ and at some point no real features are discernible.

2.1.4. Two-colour experiments : An interesting situation arises[12] when the ionization step is driven almost exclusively by one laser while another laser causes the quiver motion, the two steps depending oppositely on laser frequency. This forms the basis of most two-colour experiments. A low-frequency laser can be utilized for a large quiver amplitude generation but it will be inefficient in ionizing because of the large number of photons required. An essential requirement for the separation of ionization from the above-threshold step appears to be that the virtual inter-

mediate state in the high-frequency ionization should stay well away from the threshold. When the AC-Stark shifts are to be measured in the ionization potential for two-colour experiments, more difficulties are encountered. However, the advantage of two-colour experiments is that one has more ways to tune the experiment and thus disentangle the ionization and above-threshold steps. The two-colour experiments that use exact harmonics are more informative.

2.2. Atomic stabilization :

The non-perturbative behaviour of atomic electrons associated with ATI-type effects changes significantly if both the laser intensity and frequency are increased. At intensities above 10^{17} W cm^{-2}, some unexpected results such as stabilization and localization of the electron density may be observed. In other words, the ionization rate may be suppressed when an atom experiences such a strong field, i.e. there may be a decrease of ionization probability with increase of laser intensity. Neither localization nor stabilization appear to be fully confirmed experimentally, but theoretical studies (see later) indicate that these are possible using new-generation lasers with ultra-short pulses.

Supercomputer simulations reveal that strong-field stabilization does indeed occur[6]. At superintense fields (in excess of 1 a.u.), the laser captures the electron in the field of its wave shaking it back and forth with wave-like vibrations (light behaving more like a wave than a particle). The extent of the electron's oscillation becomes so large that the atom's wavefunction distorts into a distribution with two well-separated peaks (a bi-local form). This bi-local form[6] is maintained as laser intensity increases, thereby reducing the ionization probability; the atom is now said to be 'stabilized' in this new configuration. Another way of viewing stabilization is through Kramers-Henneberger (KH) coordinate frame in which nuclear motion is followed keeping the electron stationary. In the KH frame, at normal fields, the nucleus oscillates along the polarization axis of the laser field and bumps the electron every time it goes past, eventually resulting in ionization. As the intensity is increased, this swinging motion becomes so high that the wavefunction becomes more localized away from the nucleus, thus decreasing ionization probability.

Recently, it has been shown through computations, using correlated Hylleraas-type wavefunctions, that a doubly excited autoionizing state of He atom can be stabilized in the presence of KrF laser at laboratory field intensities[13]. This kind of stabilization results from the abrupt suppression of the dynamical electron correlation as the field intensity is increased beyond a critical value. Due to the dynamical electron correlation the He $(2s)^2$ state autoionizes spontaneously even in the absence of the field. When exposed to high intensity laser fields, the autoionization phenomenon competes with photoionization. The total ionization rate is increased and is nearly equal to the sum of the photoionization and autoionization rates. Indeed, an atom in a strong laser field may be viewed as a diatomic molecule in a field-free space with 'internuclear' distance growing with the field strength.

2.3. High harmonic generation (HHG) :

When an atom is irradiated by a high-intensity laser beam, it can emit high-order harmonics which are odd multiples (in the dipolar approximation) of incident laser frequency. The odd harmonics (coherent 0.5 keV X-ray emission corresponding to >400th order from He driven by a 5 fs laser pulse has been detected at present[14,15]) are emitted because atoms have inversion symmetry. The harmonics intensities present a characteristic distribution. After the expected rapid decrease from the first order, there is a long plateau which ends with a sharp cut-off (Fig. 2). This behavior is surprising for several reasons. First, it indicates that the higher order harmonics, e.g. the 29th harmonic, become as probable as the 5th harmonic. Therefore, a weak-field picture successfully employed for non-resonant nonlinear optical phenomena can no longer apply here. At the

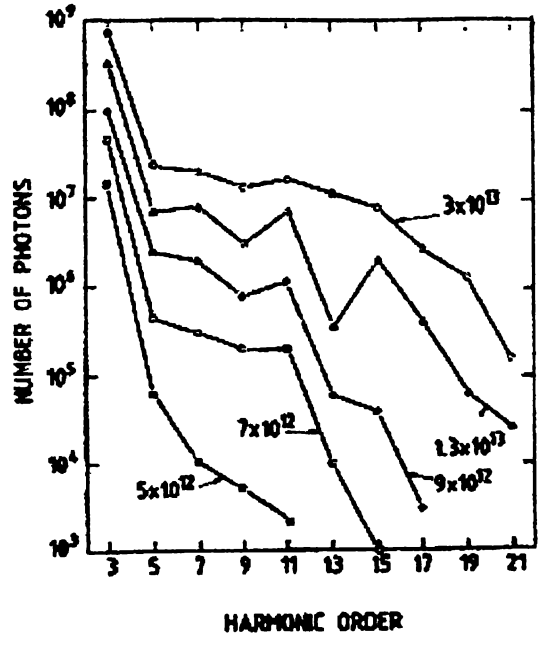

Fig. 2. Experimental HHG spectra of xenon at 1064 nm for laser intensities from 5×10^{12} to 3×10^{13} W cm^{-2}. The number of photons is plotted as function of harmonic order on a semilogarithmic scale. It may be noted that the plateau begins to appear at high intensities. (Reproduced by permission from A. L'Hullier, L. A. Lompré, G. Mainfray and C. Manus, p. 147 in Ref. 1).

lowest intensities (~10^{12} W cm^{-2}), the harmonic signal decreases with the order. As the intensity increases, a plateau appears, followed by an abrupt cut-off. The length of the plateau increases as the laser intensity increases, up to the intensity at which the medium becomes ionized with a probability close to unity, which occurs above 10^{13} W cm^{-2}. Beyond this saturation intensity, the signal increases much less rapidly than before, the distribution becomes smoother and the maximum observable harmonic order remains constant. In fact these features are common to any such strongly driven nonlinear system.

In strong fields, harmonics are created in a medium that can be partly ionized during the interaction. Free electrons can change the propagation of incident beam considerably. When their motion becomes relativistic, in very strong fields, they can produce harmonics. A plasma created by multiphoton ionization of a gas in very strong fields can generate odd, even and half-integer harmonics. Ions[16] have a higher ionization potential than atoms and one could expect ions to produce more harmonics with a longer plateau but in a less efficient manner than atoms since the potential becomes deeper and the system less polarizable. HHG from a free atom is limited by the nonlinear response of the target atom. Clusters of atoms form a new type of nonlinear medium for photoemission study in an intense short laser pulse in which a microplasma is formed which holds together inertially even when heated by electrons during the short pulse excitation. In the stabilization regime ($I > 10^{17}$ W cm^{-2}), the HHG spectra are found to contain all harmonics with a long plateau but no clear cut-offs (see later). HHG from simple molecules has also been investigated[17,18]. It was found that the harmonic yield depends strongly on the static polarizability of the species and weakly on the mass as well as ionization potential. A further limitation on HHG in molecules is that at intensities associated with efficient harmonic generation in atoms, i.e. intensities above 10^{14} W cm^{-2}, the molecules tend to 'explode' (coulomb explosion). This is due to the coulomb repulsion between the dissociated fragments. New techniques are being developed to study molecules under strong laser pulses.

2.3.1. Intensity dependence of HHG : Numerous experiments have performed for harmonic intensity dependence[19,20]. All of these present a 'knee'- or 'shoulder'-like structure; the number of photons first increases rapidly, then saturates in a way similar to the number of ions in a multiphoton ionization experiment, when saturation intensity is reached. The intensity at which the harmonic signal saturates is exactly the same as for multiphoton ionization. Thus, multiphoton ionization of an atom limits its harmonic generation. The main reasons are the depletion of the medium generating harmonics and breaking of phase matching owing to the presence of free electrons in the medium. When the medium becomes ionized, the harmonics conversion efficiency decreases although ions could also generate harmonics albeit with low response.

Indeed, both the HHG and ATI are intimately related phenomena. HHG can be viewed as a three-stage process[21] : ATI followed by a continuum electron propagation in a laser field and subsequent stimulated recombination back into the initial state. Since in the harmonic generation process, an active electron ends up in the initial bound state, it is appealing to represent it as ionization followed by recombination. This mechanism of electron-core interaction is known as atomic antenna, which is missing in Keldysh's theory of multiphoton ionization.

2.3.2. Ponderomotive effects : The harmonic generation has been observed in the regime where the ponderomotive energy is less than the ionization energy. A large number of harmonics can also be generated even when ponderomotive energy is much higher than the ionization energy provided that the laser intensity is at the ionization threshold. The region where the electric field and the tunnelling ionization rate are peaked, the free-electron density increases sharply, representing a plateau. This time-dependent density shows a high harmonic content. A large number of odd harmonics (for centrosymmetric systems in the dipolar approximation) is generated coherently along the laser field because the current density that produces the radiation is the product of the time-dependent free-electron density and the quiver velocity proportional to the electric field.

2.3.3. Coherence properties : phase matching : Coherence properties are of fundamental interest for studying the dynamics of the harmonic generation process. A characterization of coherence properties of the source is required for applications such as interferometry and spectroscopy in the XUV region[22]. The concepts of spatial and temporal coherence are important in phase characterization of harmonics. In general, the concept of coherence is related to the stability and/or predictability of phase. Spatial coherence describes the correlation between different harmonic signals at different points in space. The temporal coherence describes the correlation or predictable relationship between harmonic signals observed at different moments of time. The temporal coherence gives information about time-dependence of the harmonic radiation[23].

The behaviour of the atom-laser system in the intensity

regime needed for HHG is non-perturbative. The high-order harmonics do not scale perturbatively with intensity. Phase matching is responsible for this kind of behaviour, which can be achieved in strong fields. The intensity dependence of phases of harmonics is much stronger in the plateau than in the cut-off region, indicating an improved phase coherence of harmonics in the cut-off, a generic property of the interaction of strong laser fields with nonlinear systems. The presence of free electrons is responsible for the breakdown of phase matching, thus altering the harmonic conversion efficiency. The free electrons have a significant effect on the refractive index at a harmonic frequency, thereby inducing a large positive phase mismatch between the generated beam and its driving polarization, and hence reducing conversion efficiency.

2.3.4. Harmonic cut-off: Based on different experimental results, an empirical law has been proposed (theoretically justified later) for the cut-off in harmonic order, approximately given by

$$n \approx U_I + 3.17\ U_p/\omega_L \qquad (4)$$

where U_I is the ionization potential of the target atom, U_p the ponderomotive potential and ω_L the frequency of laser field. This relation is found to be independent of the form of potential. In some experiments, the cut-off was measured to be $U_I + k\ U_p$, $1 \le k \le 3$. This departure may be due to the short wavelength and tight focussing limit used in these experiments which made propagation effects very influential and lowered the harmonic cut-off. The larger plateau is produced by increasing U_p, i.e. using longer wavelength or higher intensities as well as increasing U_I by employing ions[24].

The 'recollision model' of harmonic generation successfully explains the cut-off[25,26] relation, eq. (4). An atom interacting with an intense laser field may be approximately described as having only one active electron and only one bound state. The electron, experiencing the sum of the coulomb field and the intense laser field, can tunnel through the barrier formed by the resulting potential and subsequently be accelerated by the laser field. For a linearly polarized laser field, the electron may be driven back towards the core and then recombine with it. While decaying to the ground state, it leads to the emission of a high-energy photon. In other words, as the atom is perturbed, it ionizes and becomes a source of electrons having 'zero' initial velocity in the laser field. For the generation of a particular harmonic, there are several possible trajectories through which the electron returns to the core with the correct energy. The electron trajectories depend sensitively on the phase of the laser field. Some of these trajectories result in ionizations while others correspond to returning to the nucleus. An electron with zero initial velocity can return to the nucleus with maximum energy equal to approximately 3.17 U_p. Recent developments in the HHG have concentrated on the ways to increase the harmonic cut-off. This has led to two approaches : (i) increasing laser intensities to above saturation levels so that harmonic generation from ions become important; this increases the operative U_I and (ii) utilizing superintense fields where stabilization becomes important so that, in effect, U_p can be increased without the prospect of severe depletion[27].

3. Theoretical treatments

At intensities less than 10^{13} W cm^{-2}, perturbation theory is adequate for describing the phenomena observed in atom-laser interaction. Above this intensity, as discussed in Section 2, non-perturbative behaviour begins to manifest itself. Spectra like those of Fig. 1 at $I < 10^{12}$ W cm^{-2} are largely accounted for by perturbation theory. Each of the peaks in the spectrum corresponds to a process of different order[28]. According to the perturbation theory, as employed in the initial understanding of MPI, the total ion yield Δ obeys the power law,

$$\Delta = \alpha\ I^N \qquad (5)$$

where the order of nonlinearity N is equal to the minimum number of photons that have to be absorbed to produce ionization. The lowest-order perturbation theory (LOPT) had successfully explained the earlier observed MPI. In this theory, the perturbed wave function Ψ_N for each order can be obtained from the previous order Ψ_{N-1} as

$$\Psi_N = (\varepsilon_n - \mathcal{H})^{-1} \hat{O}_D\ \Psi_{N-1} \qquad (6)$$

where $\hat{O}_D$ is the dipole operator representing the action of the perturbing field. The resolvent operator $(\varepsilon_n - \mathcal{H})^{-1}$ gives the response of the atom ($\mathcal{H}$ is the hamiltonian), taken at an energy ε_n that is n photons above the initial state. Strictly speaking, since the spectrum in Fig. 1 clearly shows peaks of higher order than the minimum N required to obtain ionization, it violates LOPT, although each of the peak amplitudes depends on the laser intensity with an order of nonlinearity equal to the corresponding number of photons, N, N+1, N+2. The theory describes each of the peaks satisfactorily, but up to a lowest order, indicating the rapid convergence of perturbation series at low intensities. The spectrum at intensity $\geq 10^{13}$ W cm^{-2} corresponds to a quite different physical state, with dominant higher orders, making LOPT quite inadequate. Although the Nth-power dependence of the total yield can still remain valid, the peak amplitudes at these laser intensities depend on the intensities with orders of nonlinearity considerably lower than the number of photons needed to reach the peak. This is due to the fact that with increase of laser intensity, the

different final states become strongly coupled, with the atom being driven up and down between continuum states many times during the ionization process. Therefore, the laser-induced perturbations of the resonant states, the AC-Stark shifts, have to be taken into account.

While it is clear that non-perturbative approaches are needed, these are rather difficult because no analytical solution of the Schrödinger equation for an atom in a time-varying field is known. Before proceeding to various non-perturbative approaches, we describe briefly how harmonic spectra and other physical quantities are calculated. The source of harmonic generation is the polarization of the medium induced by laser field and is given by

$$P(t) = \hat{A}_D \, d(t) \tag{7}$$

where $\hat{A}_D$ is the atomic density and $d(t)$ is the atomic dipole moment defined as expectation value of the dipole operator (er)

$$d(t) = \langle \Psi(t) | er | \Psi(t) \rangle \tag{8}$$

The commonly used method for harmonic spectrum calculation is to simply take the modulus square of the fourier transform of the atomic dipole moment defined as

$$|d(\bar{\omega})|^2 = |(1/T) \int_0^T dt \; e^{-i\bar{\omega}t} \; d(t)|^2 \tag{9}$$

However, it is more reliable to use acceleration through Ehrenfest's theorem[29],

$$\ddot{a}(t) = d^2/dt^2(\langle r \rangle) = \langle -(\partial H_{int}/\partial r) E_f \rangle; \; H_{int} = V(r) + E_f(t) \tag{10}$$

where H_{int} is the interaction part of the Hamiltonian and E_f the contribution due to the laser electric field. This is because of the large final dipole moment produced during the laser pulse particularly when significant amounts of ionization occur. Another observable of interest is the ionization probability given by

$$P_I(t) = 1 - \sum_{n=1}^{N} |\langle \phi_n | \Psi(t) \rangle|^2 \tag{11}$$

where N is the total number of bound states, ϕ_n and $\Psi(t)$ refer to the field-free bound states and time-evolved wavefunction, respectively. The ionization probability is the amount of population which is in the continuum states.

We now describe the non-perturbative approaches employed by various workers to study the atom-laser interaction.

3.1. Time-independent non-perturbative approaches :

3.1.1. Floquet and related approaches : This approach[30] involves the reduction of time-dependent Schrödinger equation (TDSE) to a system of time-independent (TI) coupled differential equations for the floquet component of the wave function,

$$|\Psi(t) = e^{-iEt/\hbar} \sum e^{-in\omega t} | \; \Psi_n \rangle = e^{-iEt/\hbar} \; \phi(r,t) \tag{12}$$

with a periodic function ϕ of frequency ω for the periodic interaction $V(t)$, given by

$$V(t) = V_+ e^{-i\omega t} + V_- e^{i\omega t} \tag{13}$$

where V_+ and V_- refer to time-independent counterparts. In eq. (12), E is the quasi-energy. The set of TI coupled equations is given by

$$[E + n\hbar\omega - H_a] | \Psi_n \rangle = V_+ | \Psi_{N-1} \rangle + V_- | \Psi_{N+1} \rangle \tag{14}$$

where H_a is the atomic Hamiltonian. These TI coupled equations have solutions with complex quasi-energies E representing the decaying states,

$$E = E_0 + \delta - i\beta/2 \tag{15}$$

where E_0 is the unperturbed energy of the state, δ the Stark shift, β the width of the decaying state and represents ionization rate. The essence of this approach lies in the stationary solutions of Schrödinger equation to describe the distortion and decay of the atom in the laser field.

For floquet-type calculations, R- and S-matrix[31] approaches have been extensively employed. In general, the floquet calculations show good agreement with the lowest energy peaks in the spectrum. The main advantage of this approach is that it is, in principle, applicable to any atom. But the disadvantage is that the calculations do not account for the full time-dependence of the incident laser pulse. Another physical limitation is the idealization that the field has constant amplitude whereas, in practice, superintense laser fields are generated in short pulses. But this may be resolved by introducing a prior time-dependence in the constant amplitude results, thereby permitting the description of the system adiabatically from an initial unperturbed state.

3.1.2. Volkov final state methods : Solutions of Schrödinger equation for a free electron in a plane wave electric field are known as Volkov states[32,33]. The Volkov states are plane waves with an oscillating phase, which depends on the vector potential. An ionizing electron is assumed to make a transition between the initial bound coulombic wavefunction and the final free-electron Volkov state. Utilizing the Volkov states as possible final states of the electron, Keldysh[13], Faisal[34] and Riess[35] (KFR) had developed an approach by employing field-free eigenstates as the initial wavefunctions, connecting them with an interaction. A set of coupled first-order differential equations is solved to obtain the solution, which is the time-dependent expansion coefficient of the bound and Volkov states. Although such approaches enjoy their own success, they neglect the laser-induced level shift, the AC-Stark shift and the effect of the ionizing electron. These effects become more important at higher laser intensities. Furthermore,