ATOMS IN INTENSE LASER FIELDS

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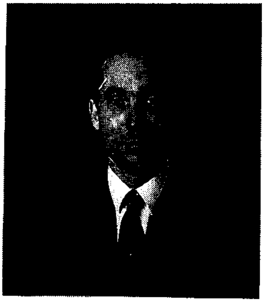

ABSTRACT

A review is given of recent progress made in studying the interaction of atomic systems with intense laser fields. Following a survey of multiphoton ionization, harmonic generation and laser-assisted electron-atom collisions, several non-perturbative theories of laser-atom interactions are discussed. Future developments of this rapidly expanding area of atomic physics are also considered.

1. INTRODUCTION

The study of atoms interacting with intense laser fields is one of the most rapidly growing areas of atomic physics. Advances in laser technology have led to the discovery of new phenomena in laser-atom interactions, mainly via the investigation of multiphoton processes. Lasers are now available, which are capable of producing oscillating electric fields of strength $\mathcal{E}_0$ larger than the atomic unit of electric field $e/a_0^2 \simeq 5.1 \times 10^9$ Vcm^{-1}, the corresponding intensity being $I \simeq 3.5 \times 10^{16}$ Wcm^{-2}.

In this article we shall review some recent progress made in studying the interaction of atoms and ions with intense laser fields. We begin in Section 2 by giving a survey of several important multiphoton processes: multiphoton ionization, harmonic generation and laser-assisted electron-atom collisions. In Section 3, we discuss non-perturbative theories which have been developed to analyze these phenomena. Finally, in Section 4, we examine some possible future developments . Comprehensive reviews of the physics of atoms in intense laser fields have been written recently by Burnett et al [1)] and Joachain [2)]. Detailed articles covering various aspects of the subject may also be found in a volume edited by Gavrila [3)].

2. MULTIPHOTON PROCESSES

The multiphoton phenomena occuring in laser-atom interactions are conveniently divided in two classes: laser-induced and laser-assisted processes. Laser-induced processes can only occur significantly in the presence of the laser field, while laser-assisted processes can take place in the absence of the laser field, but are modified in its presence. In what follows we shall consider two types of laser-induced processes: multiphoton ionization of atoms and ions and harmonic generation. Laser-assisted processes, on the other hand, will be illustrated by examining electron-atom collisions in the presence of a laser field.

2.1 Multiphoton ionization of atoms and ions

We begin by considering the multiphoton (single) ionization (MPI) reaction

$$n\hbar\omega + A(i) \rightarrow A^+(f) + e^- \tag{1}$$

where $A(i)$ is an atom (ion) in state i and $A^+(f)$ the corresponding ionized system in state f. This process was first observed in 1965 by Voronov and Delone [4)], who used a ruby laser to induce seven photon ionization from the ground state of xenon, and by Hall et al. [5)], who recorded two-photon ionization from the negative ion I^-. Important results were then obtained by several groups concerning the dependence of the ionization yields on the laser intensity, the resonant enhancement of MPI and multiphoton multi-ionization .

A crucial step forward in the understanding of MPI was made when expriments detecting the ionized electrons were performed. Thus Agostini et al [6)] , who measured the energy of the photoelectrons produced by the MPI reaction, discovered that the ejected electron could absorb photons in excess of the minimum required for MPI. The study of this excess-photon ionization, currently known as "above threshold ionization" (ATI) has been one of the central themes of multiphoton physics in recent years. The ATI photoelectron energy spectra consist of several peaks, separated by the photon energy $\hbar\omega$. As the laser intensity increases, the number of these peaks becomes larger, the ratios of their intensities do not follow the predictions of perturbation theory , and the lowest order peaks are reduced and eventually suppressed. The reason for this peak suppression is that the energies of the atomic states are shifted in the presence of a laser field. For low frequencies (e.g. the Nd-YAG laser, for which $\hbar\omega = 1.165$ eV) the dynamic Stark shifts for the lowest bound states (in particular the ground state) are small in magnitude. On the other hand, the shifts of the Rydberg and continuum states are essentially given by the electron ponderomotive energy U_p, which is the kinetic energy due to the quiver motion of the electron in the laser field, averaged over a laser cycle. This ponderomotive energy is given by

$$U_p = \frac{2\pi e^2 I}{mc\omega^2} \tag{2}$$

where m is the electron mass, I is the laser intensity and ω its angular frequency. For example, in the case of the Nd-YAG laser, $U_p = \hbar\omega = 1.165$ eV when $I \simeq 10^{13}$ Wcm^{-2}. Since the energies of the Rydberg and continuum states are shifted upwards relative to the lower bound states by about U_p, there is a corresponding increase in the ionization threshold. If this threshold increase is such that $E_i + n\hbar\omega - U_p < 0$, where E_i is the unperturbed (field free) energy of the initial state, then the peak in the ATI spectrum corresponding to ionization by n photons will be suppressed. Interesting effects in ATI spectra due to the influence of the laser pulse duration have also been reported [7,8].

When the ponderomotive energy U_p is much larger than the ionization potential, ionization occurs by the tunneling mechanism, i.e. the electron tunnels through the barrier formed by the atomic potential and the laser electric field [9], provided that the field is of low frequency and not so strong that field ionization occurs. In this case the regular structure of ATI peaks disappears. Field ionization [10] takes place when the laser intensity reaches a critical value I_c (which is about 1.4×10^{14} Wcm^{-2} for the ground state of atomic hydrogen) such that the saddle point of the barrier is so lowered that the electron can "flow over the top"; in this regime the atom ionizes in about one orbital period.

If both the intensity and the frequency of the laser field are large, it is predicted that the atom should become stable against ionization, and that the ionization rate should decrease as a function of the intensity in the high frequency, high intensity regime [11,12]. This "stabilization" of atoms at super-intensities is currently attracting considerable attention.

2.2 Harmonic generation

Atoms irradiated by laser light can emit radiation, whose frequency is an odd multiple $N\omega$ of the laser frequency ω because of parity conservation . This harmonic generation process has been the subject of active investigations for many years. Third harmonic generation in a rare gas medium was observed for the first time in 1967 by New and Ward [13] while fifth - and higher - order harmonics were seen a few years later. Recently, it has been shown [14-16] that very high (odd) harmonics could be generated when atoms interact with intense laser pulses. For example, in the experiments of L'Huillier and Balcou [16], performed with a Nd-YAG laser of photon energy $\hbar\omega = 1.165$ eV, the harmonic order $N = 133$ was observed in the case of neon. High order harmonic generation has attracted considerable interest as a potential source of short pulse, high frequency coherent radiation. The harmonic intensity distribution consists of a rapid decrease over the first few harmonics, followed by a plateau of approximately constant intensities and a cut-off. This general structure is in good qualitative agreement with calculated single-atom spectra, indicating that propagation effects in the medium play a minor role, or affect all the harmonics in the same way. The existence of a plateau in the harmonic intensity distribution is a non-perturbative effect which appears to be a very general characteristic of strongly driven non-linear systems.

2.3 Laser-assisted electron-atom collisions

Electron-atom collisions in the presence of laser field (also called free-free transitions) have attracted a great deal of attention not only because of the importance of these processes in applied areas (such as plasma heating), but also in view of their interest in fundamental atomic collision theory. In the first experiments performed by Andrick and Langhans [17] with low-intensity lasers, the transfer of only one photon between the electron-atom system and the laser field was observed. Subsequently, however, several experiments [18-20] have been carried out in wich the transfer of several photons has been observed in laser-assisted collisions of the type

$$e^- + A(i) + l\hbar\omega \rightarrow e^- + A(f), \tag{3}$$

where positive integer values of l correspond to photon absorption (inverse bremsstrahlung), negative integer ones to photon emission (stimulated bremsstrahlung) and $l = 0$ to a collision process without net absorption or emission of photons but in the presence of the laser field.

It is interesting to note that the observation of multiphoton phenomena in free-free transitions requires much lower intensities than for laser-induced processes. For example, significant rates for the emission or absorption of 10 laser quanta have been observed [19] in laser-assisted "elastic" electron-argon collisions at a laser intensity of about 10^8 Wcm^{-2}.

3. NON-PERTURBATIVE THEORIES

Most of the early theoretical treatments of multiphoton processes relied on the use of perturbation theory. However, in the case of strong laser fields, non-perturbative methods are required. We shall discuss below two of these methods: the Floquet theory and the direct numerical integration of the time-dependent Schrödinger equation.

3.1 Floquet theory

Although more general laser fields can be considered (for example "two-color " fields) we shall assume that we are dealing with a laser field wich is treated classically as a spatially homogeneous, monochromatic electric field of angular frequency ω . Then, in both the length or the velocity gauge, the Schrödinger equation describing the atomic system in the presence of the laser field is

$$i\hbar\frac{\partial}{\partial t}\,|\Psi(t)>=[H_{at}+H_{int}(t)]\,|\Psi(t)> \tag{4}$$

where H_{at} is the field free atomic Hamiltonian and the laser-atom interaction term $H_{int}(t)$ can be written in the form

$$H_{int}(t)=H_{+}e^{-i\omega t}+H_{-}e^{i\omega t} \tag{5}$$

where H_+ and H_- are time-independent operators. The Hamiltonian of the system, $H(t) = H_{at} + H_{int}(t)$, is periodic , i.e. $H(t+T) = H(t)$, where $T = 2\pi/\omega$. The Floquet method can then be used to write the state vector $|\Psi(t)>$ in the form

$$|\Psi(t)>=e^{-iEt/\hbar}\;|F(t)> \tag{6}$$

where the "quasi-energy" E does not depend on time and $|F(t)>$ is periodic in time, with period T, so that it can be expressed as the Fourier series

$$|F(t)>=\sum_{n=-\infty}^{+\infty}e^{-in\omega t}\,|F_n> \tag{7}$$

The $|F_n>$ are called the harmonic components of $|F(t)>$. Using equations (4)-(7) one obtains for the harmonic components $|F_n>$ the time-independent infinite system of coupled equations

$$(E+n\hbar\omega-H_{at})|F_n>=H_{+}|F_{n-1}>+H_{-}|F_{n+1}> \tag{8}$$

These equations, together with appropriate boundary conditions, form a problem which in general must be solved by keeping only a finite number of harmonic components, i.e. by truncating the sum on n in eq. (7) .

Two non-perturbative methods have been used successfully to solve the coupled equations (8). The first one, wich in practice is limited to one-electron systems, is the Sturmian-Floquet method[21]. In this approach, each harmonic component in position space $F_n(\mathbf{r}) \equiv <\mathbf{r}|F_n>$ is expanded on a discrete basis set consisting of spherical harmonics and complex Sturmian radial functions. This method has been applied extensively to study multiphoton ionization and harmonic generation in atomic hydrogen. A review of this work has been given by Potvliege and Shakeshaft[22]. We note in particular the good agreement between the Sturmian-Floquet calculations of Dörr et al.[23] and the experimental data of Rottke et al.[24] for multiphoton ionization of H(1s).

The second non-perturbative method is a new approach - the R-matrix-Floquet theory - which has been proposed recently [24] to analyze multiphoton processes. It combines the powerful R-matrix theory with the Floquet method to treat multiphoton ionization, harmonic generation and laser-assisted electron-atom collisions in an unified way. It is completely ab initio and is

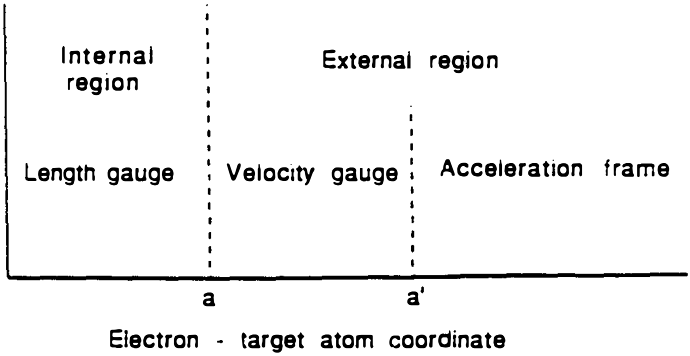

Figure 1: Partitioning of configuration space in the R-matrix-Floquet theory.

applicable to an arbitrary atom or ion. According to the R-matrix method, configuration space is subdivided into two regions (see Fig. 1). The internal region is defined by the condition that the coordinates r_i of all N electrons of the atomic system are such that $r_i \leq a$, where the sphere of radius a envelops the charge distribution of the target atom(ion) states retained in the calculation. Hence in this region exchange effects involving all N electrons are important. The external region is defined so that one of the N electrons lies on or outside the sphere of radius a ,while the remaining $(N-1)$ "target" electrons are confined within this sphere. Thus in this region exchange effects between this one electron and the remaining $(N-1)$ "target" electrons are negligible. The Schrödinger equation (4) is solved in these two regions separately by using the Floquet method, and the solutions are then connected on the boundary at $r = a$. In the internal region it is convenient to use the length gauge since in this gauge the laser-atom coupling tends to zero at the origin. In the external region it is advantageous to use the velocity gauge out to a radius a' which may extend to infinity. In certain cases it is useful to transform at $r = a'$ to the Kramers acceleration frame which enables simple asymptotic boundary conditions to be defined. The R-matrix-Floquet theory has already been applied with great success to analyze multiphoton processes in atomic hydrogen[25,26],in the two-electron systems H^- and He[27] and in more complex atoms such as neon and argon[28]. Branching ratios into the ATI channels have been obtained, as well as angular distributions for the photoelectrons. Atomic stabilization at high frequencies and high intensities has been studied, and novel non-perturbative correlation effects[27] and laser-induced degeneracies[28] have been found.

3.2. Numerical integration of the time-dependent Schrödinger equation

One of the major non-perturbative techniques which has been used in recent years to study atoms in strong laser fields is the direct, numerical solution of the corresponding time-dependent Schrödinger equation. This approach, which is particularly useful for very short laser pulses (where the Floquet method is not applicable), has been used in two different ways. Firstly, "numerical experiments" of multiphoton processes have been conducted in studies using one dimensional potentials[29]. Secondly, numerical methods have been used to solve the time-dependent Schrödinger equation in the dipole approximation for "realistic" (three-dimensional) atoms in pulsed laser fields[30]. Most of these computations have been single electron calculations that are "exact" for hydrogenic systems, but only approximate for atoms or ions with more than one electron. Very recently, atomic hydrogen in a super-intense, high frequency field has been studied beyond the dipole approximation[31] by solving numerically the time-dependent Schrödinger equation. In addition, the coupling of the magnetic field to the spin of the electron has also been included by solving the time-dependent Pauli equation. Adiabatic stabilization was obtained at high intensities, and corrections to the dipole approximation were found to modify the ionization probability only slightly.

4. FUTURE DEVELOPMENTS

Lasers will soon become available, which can deliver ultra-intense (10^{20} W cm^{-2}) and ultra short (femtosecond) pulses. The study of laser-atom interactions under these conditions will require the development of non-perturbative theories taking fully into account relativistic effects. New phenomena will also become accessible to investigation, such as the excitation and ionization of inner shell atomic electrons, electron-positron pair production and multiphoton Compton scattering, leading to very high harmonic generation. Clearly, this area of research holds great potential for future developments.

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