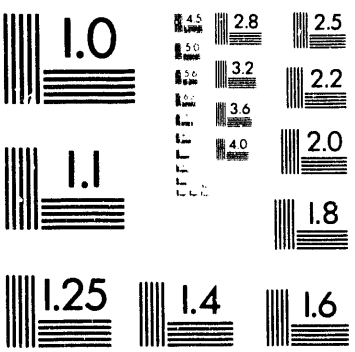
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Quantum and Quasi-Classical Analyses of High Order Emission Processes

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Abstract

High-order harmonic generation from atoms and ions cuts off in accord with the simple rule: $E_c \sim I_p + 3U_p$. This cutoff rule as well as the *lack of a cutoff* in high-order electron energy spectra can be understood via a simple extension of existing two step quasi-classical models.

Introduction

Recent quantum mechanical calculations have shown that an atom in a strong laser field of intensity I and frequency ω will radiate high-order harmonics up to a maximum energy of $E_c \sim I_p + 3U_p$ [1]. This cutoff energy, which defines the end of a "plateau" of harmonics of comparable strength, scales as the ionization potential of the atom, I_p, and the ponderomotive energy U_p, given by $I/4\omega^2$. In this paper we show that the high harmonic cutoff, as well aspects of above threshold ionization (ATI), can be understood via a straightforward extension of the "Simpleman theory of ATI" proposed by van Linden van den Heuvell and Muller [2] and the tunnel ionization model of Gallagher and Corkum [3]. We label these analyses "quasi-classical" since they involve a two step process: quantum initiation (ionization) followed by classical motion. Throughout, we compare the quasi-classical

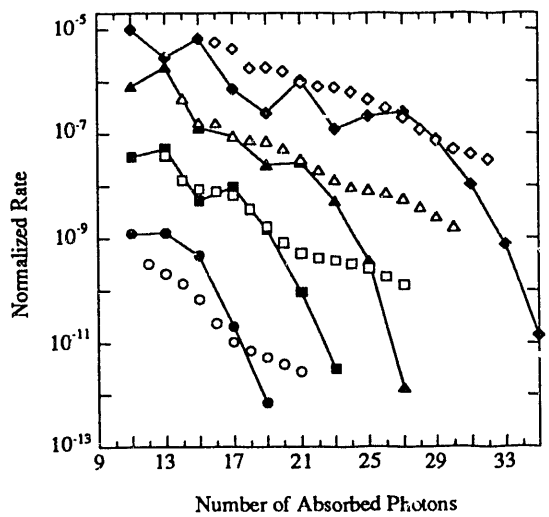

Figure 1. Single atom harmonic generation (closed symbols) and above-threshold ionization (open symbols) rates for 1064 nm and 1×10^{13} (circles), 2×10^{13} (squares), 3×10^{13} (triangles) and 5×10^{13} (diamonds) W/cm^2.

predictions with quantum calculations based upon integration of the time-dependent Schrodinger equation.

Our interest in quasi-classical models originated in our efforts to understand the results in Fig. 1. We plot photon and electron emission rates for xenon at 1064 nm and intensities between 1 and 5 x 10^{13} W/cm^2. The rates have been normalized to agree for the 17 photon processes at 3 x 10^{13} W/cm^2 and are for a single atom (see the paper of L'Huillier, *et al.*, in this volume for a discussion of propagation effects). They were obtained

by integrating the time-dependent Schrodinger equation in the single active electron approximation [4]. Of immediate note is the fact that the harmonic generation spectra all show a distinct cutoff while the electron spectra do not. Both results are in agreement with experiment [4,5].

Quasi-Classical ATI

The description of above threshold ionization spectra via quasi-classical means dates at least to the "Simpleman's theory of ATI" proposed by van Linden van den Heuvell and Muller [2]. They suggested that ATI spectra could be understood by considering a two step process. First, some portion of the electron wave function is liberated from the atom at each instant of the optical cycle. This step is necessarily quantum mechanical but may be described by perturbation theory, resonant or non-resonant multiphoton ionization, or tunnel ionization. However the electron is freed, its subsequent motion (the second step) is treated classically and any further effects of the ionic potential are ignored. The atom acts as a source of electrons that are introduced into the continuum at a time t_o.

In this simple model the cycle averaged energy of the electron depends only upon t_o (i.e., the phase of the electric field at which the electron is "born"). It is given by

$$\langle E \rangle = U_p\left[1 + 2\cos^2(\omega t_o)\right], \qquad (1)$$

where we have assumed that the initial velocity of the electron is negligible. There are two pieces: the drift energy due to acceleration by the field during the first half cycle of freedom, which has a maximum value of $2U_p$, and the quiver energy, U_p, which is converted to drift energy in the long pulse limit. The Simpleman model predicts that the maximum number of ATI photons that can be absorbed is therefore $I_p + 3U_p$. This corresponds to, for example, ~ 19 photons for 3 x 10^{13} W/cm^2 and 1064 nm. This is clearly contradicted by the quantum results in Fig. 1 and by recent experimental results [4].

Missing from this description of course is the possibility that an electron, once freed, might have a collision with the nucleus. For linear polarization this is actually quite probable since many of the classical trajectories pass near the nucleus at least once. An elastic collision that causes the electron to deviate from a straight line trajectory can convert quiver energy to drift energy in the direction perpendicular to the polarization. This gives a likely source for the high energy electrons observed. It also suggests that the angular distributions of high order ATI peaks should show additional structure compared to low order peaks. Indeed, there is evidence for this effect in the data of Wolff, *et al.* [6].

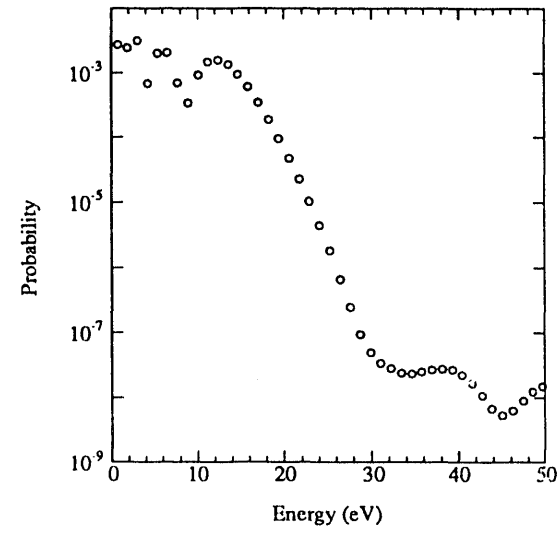

Figure 2. ATI spectrum for 1D model atom at 5 x 10^{13} W/cm^2 and 1064 nm. $2U_p$ corresponds to 10.5 eV.

As an aside, we note that one-dimensional (1D) model atoms do exhibit an ATI cutoff in accord with the Simpleman model. We show a typical case in Fig. 2 which uses the soft Coulomb potential popularized by Eberly and coworkers. The only elastic collision able to

add drift energy in 1D corresponds to back scattering. Evidently the phase space for this process is quite small, leading to a rapid decline in the spectrum beyond $2U_p$ (these calculations correspond to the "short pulse" limit).

Quasi-Classical Harmonic Generation

We can formulate an equivalent quasi-classical model for harmonic generation. The main features we seek to describe are the broad plateau of harmonics above the ionization threshold and their abrupt cutoff. Since these are most evident at high intensities, it is appropriate to think in terms of the tunnel ionization model of Gallagher and Corkum [3]. Again we assume that some fraction of the electron wave function is liberated at each instant of the optical cycle due to tunneling through the barrier created by the sum of the Coulombic attraction of the ion core and the instantaneous electric field. The subsequent electron motion is treated classically. We assert that only trajectories that lead to the return of the electron to the nucleus can cause high-order harmonic emission and that the energy that the electron has *at the time it returns to the ion* determines the energy of the emitted photon. This is distinct from the cycle averaged nature of ATI.

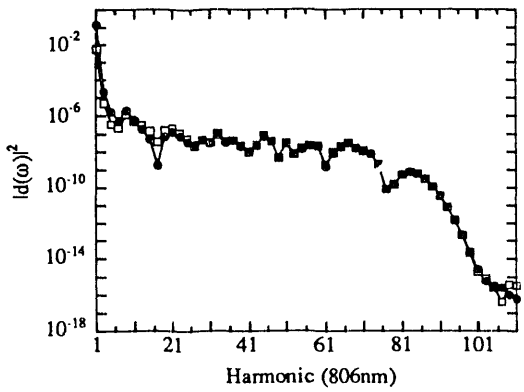

Figure 3. Exact (full symbols) and approximate (ground state transitions only, open symbols) single atom harmonic emission spectrum for neon at 6 x 10^{14} W/cm^2 and 806 nm.

Fig. 3 illustrates the fact that high harmonic emission comes from transitions to the ground state of the atom, which occur only when the excited electron is near the ion. We compare a full quantum calculation for the single atom harmonic generation spectrum to a spectrum calculated using only transitions that end in the field free ground state of the atom [7]. As the figure demonstrates, this is an excellent approximation above the ionization threshold.

A model that we might call the "Simpleman's theory of harmonic generation" can be formulated by assuming that all ionizing trajectories start at the nucleus with no initial velocity and that the effects of the ionic potential can be ignored. Integrating the classical equations of motion one finds that the maximum energy an electron can have when it returns to the nucleus is $3.2U_p$. A better treatment takes account of two factors: (i) the electron's initial position is displaced to the point just outside the instantaneous barrier (true "tunneling" initiation) and (ii) the potential substantially affects the motion of the electron. The potential also contributes the binding energy, I_p to E_c. If the initial displacement is large the return energy can be much higher than $3.2U_p$, but this condition is unlikely because the tunneling rate through the broad barrier necessary for such a displacement is extremely small. Fig. 4 shows the (tunneling) ionization rates and return energies as functions of the phase of the electric field when the electron is freed, taking full account of the atomic potential and the initial displacement. For initial phases near $\pi/2$, where the electric field is a maximum, the drift velocity is quite small and the electron can have multiple collisions with the ion. As the initial phase increases further, the initial displacement increases along with the drift velocity and only single encounters occur. We find that most of the probable

orbits (those with non-negligible ionization probabilities) fall within the limits of the Simpleman model.

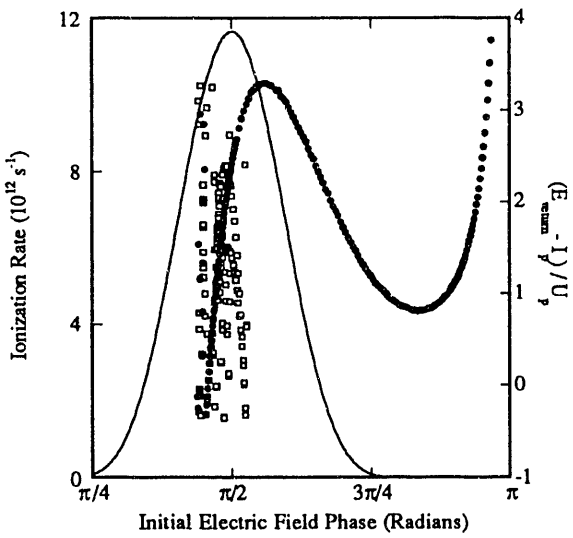

Figure 4. Tunnel ionization rate (line) and E_{return} - I_p (in units of U_p) for collisions during the first cycle after ionization (filled circles) and during the second through sixth cycles after ionization (open squares) vs. the initial electric field phase. The frequency is 0.025 a.u. and the intensity is 10^{14} W/cm^2.

When the electron returns to the nucleus it can make a one-photon transition back to the ground state. A histogram of the return energies weighted by the probability to have ionized at the phase which yields the correct return energy times the one-photon matrix element squared produces an accurate representation of the observed plateau (i.e., a relatively flat distribution) and cutoff. (Lewenstien has incorporated some of these same ideas into a quantum tunneling model with similar results. See the paper of L'Huillier, *et al.*, in this volume). The dipole matrix element varies as $\sim\omega^{-3}$, though there could be some observable structure due to the atomic potential, e.g., a Cooper minimum (See, for example, the Ar data in Fig. 3 of Ref. 1b). Of course this model is totally incapable of describing harmonics close to or below the ionization threshold since bound state effects are ignored.

Two Color Effects

The quasiclassical model yields other testable predictions. Consider, for example, the combination of a laser and its second harmonic

$$\varepsilon(t) = \varepsilon_o\left[\cos(\theta)\sin(\omega t) + \sin(\theta)\sin(2\omega t + \phi)\right] \quad (2)$$

The mixing angle θ gives the relative amounts of 1ω and 2ω light while ϕ is a (constant) phase difference between the two fields. The cycle averaged intensity is independent of θ and ϕ, while the ponderomotive potential is

$$U_p(\theta) = U_p(1\omega)\left\{\cos^2(\theta) + \sin^2(\theta)/4\right\} \quad (3)$$

Integrating the classical equations of motion for this field we find that E_c depends upon the ratio of the field strengths ($\tan\theta$) and, to a lesser extent, the phase difference between the fields. The predicted cutoffs, shown in Fig. 5, agree well with exact calculations, resulting in cutoffs as high as $\sim I_p + 5.2U_p(\theta)$. This does not result in any large enhancement of E_c, however, since $U_p(\theta)$ is a decreasing function of θ.

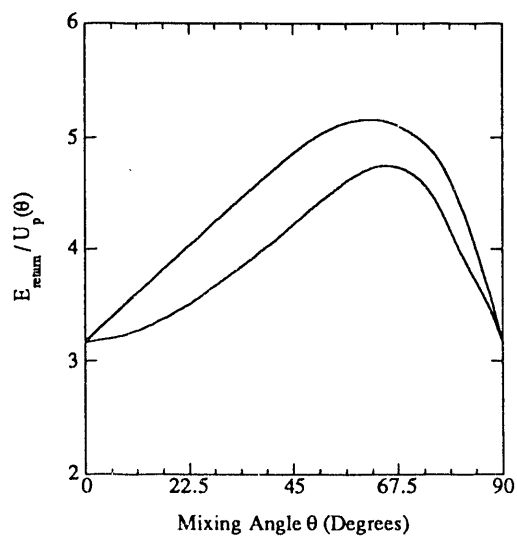

Figure 5. E_{return} in units of $U_p(\theta)$ from the classical equations of motion for the field in Eq. 2. The two curves show the maximum and minimum return energies taking account of all possible values of the relative phase ϕ.

Conclusion

We have shown that the inclusion of scattering from the ion core in two step quasi-classical models for ionization can explain both the high-order harmonic generation cutoff as well as the lack of a cutoff in long pulse ATI distributions. We note that the electron returns to the nucleus with much more energy than it had immediately following its transition to the continuum. It is the acceleration of the electron when it is far from the atom by the oscillating field which provides this additional energy. The applicability of the quasi-classical model to other high intensity effects remains an active area of research.

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