

Multiphoton ionization of hydrogen in parallel microwave and static fields: quantal and classical simulations

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Abstract

The ionization of hydrogen in strong microwave fields is a fundamental problem of atomic physics and nonlinear dynamics. When a strong static field is added parallel to a linearly polarized microwave field of comparable strength, transitions between dressed states take place simultaneously at several resonance frequencies. We present classical and quantal simulations of this intricate problem which reproduce its distinctive features very well.

1. Introduction

Understanding fundamental quantum systems in regimes of strong perturbation is a matter of wide interest in physics. One-electron systems are the most fundamental at the atomic level, and have been a source of remarkable discoveries in the past three decades. One of them, the ionization of hydrogen in a strong microwave field [1], has revolutionized the way we view the physics of highly excited atoms (see [2–4] for reviews). The interpretation of the latter problem remained a puzzle to atomic theory until its stochastic, diffusional nature was uncovered through the then-new theory of chaos [5]. In the intervening two decades, much of the research on the theory side was motivated by the recognition of the microwave ionization problem as a testing ground for quantal manifestations of classical chaos [6].

Recent experiments in which a strong static electric field is added parallel to a microwave field of comparable strength [7, 8] have once again presented atomic physicists and nonlinear dynamicists with new challenges. The detailed experiments of Koch *et al* [8] show ionization yield curves which are rich with regular oscillatory features and signatures of resonance transitions in the form of sharp dips. Transitions arise from the interplay of the strong static fields and the dressed states created by the strong microwave field. When they are treated coherently as multifrequency transitions [9] driven by a single-frequency microwave field

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between Floquet (or quasi-energy) states (QES) [10–13], many experimental features can be explained very well using an analytical, closed-form theory [14, 15]. One prominent feature of these experiments, however, remained unexplained: the rapid oscillations that are superimposed on the resonance dips [8]. Since these oscillations were suspected to be a subtle quantal phenomenon, we report detailed simulations—including the first quantal simulations of the parallel fields scenario—which agree strikingly well with corresponding classical simulations as well as existing experimental data, and provide a practical predictive tool for future experiments in this complex scenario.

2. Theory

A quantum system placed in a periodic external field can be described in terms of nonstationary QESs [11, 12] which play the same role as do stationary states in a time-independent system. This development is based on the formalism of Floquet solutions [10, 13] which form an orthonormal basis (see monographs like [16] for a general description).

Our Hamiltonian is

$$H(t) = H_1 + \hat{z}F(t) = H_1 + \hat{z}(F_s + F_\omega \cos \omega t) \quad (1)$$

where H_1 is the atomic Hamiltonian and $F(t)$ is the combined external field. Because of the cylindrical symmetry, it is natural to express its solutions in the parabolic basis. The formulation in [14] identifies the operator that creates the QES and the one that drives the transitions between them. The same theory shows the origin of the dips in the survival probability when the integer j , the number of microwave quanta involved in the resonance, is given by

$$j \equiv 3n \frac{F_s}{\omega}. \quad (2)$$

In the experiment, condition (2) selects a series of static field strengths $F_s(j) = j\omega/(3n)$ at which dips are observed in the survival probability.

3. Simulations

We performed both quantum and classical calculations to simulate the ionization of hydrogen atoms for experimentally accessible parameters. In all of the simulations, the hydrogen atoms were at an energy corresponding to an $n = 39$ state but with a completely random orientation and initial angular momentum. The field strength for the microwave and static fields were ramped on at the initial time $t = 0$ and ramped off at the final time t_f by multiplying them by the smooth function $[1 + \operatorname{erf}(t/5\tau)]/2$ for the turn-on and $[1 - \operatorname{erf}([t - t_f]/5\tau)]/2$ for the turn-off where τ is the microwave period, $2\pi/\omega$.

The quantum calculations were performed using a split operator method. The wavefunction is represented by a sum of radial functions times spherical harmonics:

$$\psi(t) = \sum_{\ell} R_{\ell}(r, t) Y_{\ell m}(\theta, \phi). \quad (3)$$

The maximum angular momenta in a typical calculation were 60–70. The Hamiltonian was split into two parts with H_1 equal to the zero-field atomic Hamiltonian and H_2 equal to the Hamiltonian from the static plus microwave field: $F(t) \cdot \hat{z}$. The propagation for one time step, δt , is accurate through order $(\delta t)^3$ and uses an implicit propagator for H_1 and an explicit propagator for H_2 :

$$\begin{aligned} \psi(t + \delta t) = & (1 - i\delta t H_1/4)(1 + i\delta t H_1/4)^{-1} \exp[-iH_2(t + \delta t/2)\delta t] \\ & \times (1 - i\delta t H_1/4)(1 + i\delta t H_1/4)^{-1} \psi(t). \end{aligned} \quad (4)$$

The Hamiltonian for the atom, H_1 , is approximated using a radial grid with a square root mesh and a Numerov approximation for the kinetic energy operator. Typically, the wavefunction was contained within a spherical region with a radius of 7000 au; a mask was used to absorb outgoing electron probability and prevent the reflection of outgoing electrons from the spherical boundary. The survival probability for a specific ℓ, m initial state is simply the norm of the wavefunction after the fields were ramped off.

The initial wavefunction was chosen to be an eigenstate of the $n = 39$ manifold. A completely random distribution would use the average of all possible ℓ, m for the initial state; this would necessitate roughly $n^2/2 \sim 750$ runs since the ionization probability for a given ℓ, m is the same as for $\ell, -m$. We found that accurate results could be obtained by putting all ℓ, m states into a random order and computing the survival probability for only the first N states: the results appeared to converge after ~ 45 initial states.

The classical calculations were performed using an adaptive step-size Runge–Kutta method. The calculations are deceptively complicated. The difficulty is in the chaotic nature of the trajectories. The trajectory was calculated so that at each time step the estimated error was less than a fixed scaled error. The estimated error was then reduced by a factor of ten. A trajectory was considered to be converged if the final energy differed by 0.02% for electrons that remained bound *or* the ionization time differed by less than five times the time step. For some initial conditions, convergence could not be achieved (the time step would be reduced below the level that round-off errors became significant) and the results from the smallest time step were used to determine whether the trajectory ionized or not. The survival probability is computed as a Monte Carlo procedure using ~ 3000 initial conditions to sufficiently reduce the statistical errors.

The distribution of initial conditions is chosen to give a completely random initial state with the requirement of being at the energy of the $n = 39$ state. The angular momentum is given by $L = \sqrt{x} \cdot 39$ where x is chosen from a flat, random distribution $0 \leq x \leq 1$. The initial radius is chosen to be the outer turning point for the given L ; random distribution in r is achieved by choosing an initial time as $t_0 - \tau_{ran}$ where τ_{ran} is chosen from a flat, random distribution between 0 and the Rydberg period. The angular part of the distribution is computed in two steps. In the first step, the initial position is in the xz plane so that $z = r \cos \theta$ with $\cos \theta$ chosen from a flat random distribution between -1 and 1 ; the initial velocity is chosen so $\mathbf{v} = v_0(\cos \theta \sin \phi, \cos \phi, -\sin \theta \sin \phi)$ where ϕ is chosen from a flat random distribution between 0 and 2π . In the second step, the initial positions and velocities are rotated about the z -axis through a random angle chosen from a flat distribution between 0 and 2π .

4. Comparison with experiment

The condition (2) gives the locations of the dips in the survival probability in terms of the static electric field $F_s(j) = j\omega/(3n)$. For the experimental conditions presented in figure 6 of [8] and the maximum microwave amplitude 319 V cm^{-1} , our paper [14] gave $F_s(1) = 47 \text{ V cm}^{-1}$, and $F_s(2) = 94 \text{ V cm}^{-1}$, in good agreement with the experimental locations of the experimental dips.

Now we concentrate on the dip at $F_s(1) = 49 \text{ V cm}^{-1}$ by simulating the survival probability around this value for small steps of the field strength. The results appear in figure 1. This first quantal simulation for parallel fields reproduces very well the well known slow oscillations [8] of the survival probability. It is also interesting that the quantal simulations practically coincide with the classical simulations and the experimental data in the vicinity of the dip. The agreement continues to be satisfactory away from the resonance between the classical and quantal simulations. The simulations are in qualitative agreement with the slow

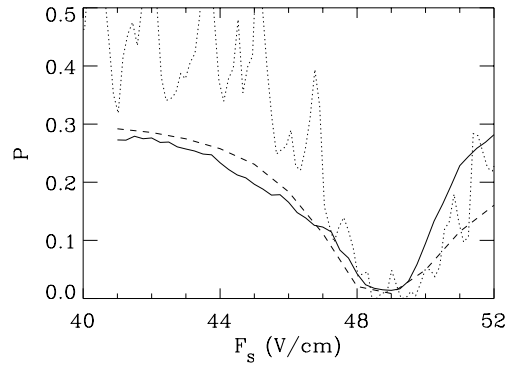


Figure 1. Survival probability, P , for $n = 39$ states near a resonant dip as a function of the static field strength F_s . The solid and dashed curves show our quantal and classical simulation results, respectively. The dotted curve is the experimental findings (see [8], figure 6). The maximum microwave amplitude, F_ω , is 319 V cm^{-1} , the frequency is 8.1 GHz and the microwave field is turned off at $t_f = 150$ periods.

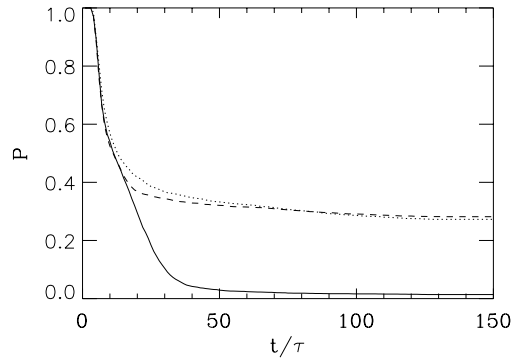


Figure 2. The survival probability, P , as a function of time in units of the microwave period, τ . The microwave parameters are the same as in figure 1. The static field strengths are below the resonance (41 V cm^{-1} , dotted curve), on resonance (49 V cm^{-1} , solid curve) and above resonance (52 V cm^{-1} , dashed curve).

dependence of the experimental survival probability on the static field. However, the quantal simulations do not yield the rapid oscillations [8] of the survival probability, the origin of which have been subject of speculation. Our simulations eliminate the hypothesis that these rapid oscillations are a fundamental quantal result.

The theoretical findings displayed in figure 2 will have to await future experiments: these are time-resolved simulations of the survival probability for various field strengths near the resonance; the results are for static fields below resonance (41 V cm^{-1} , dotted curve), on resonance (49 V cm^{-1} , solid curve) and above resonance (52 V cm^{-1} , dashed curve). For times $t \lesssim 17\tau$

- (i) the static field does not play a role since the survival probability does not depend on the field strength during this period,
- (ii) about 60% of hydrogen atoms (initially in $n = 39$) are ionized during this time.

During later times, the ionization shuts down for all practical purposes except when the static field satisfies the resonance condition.

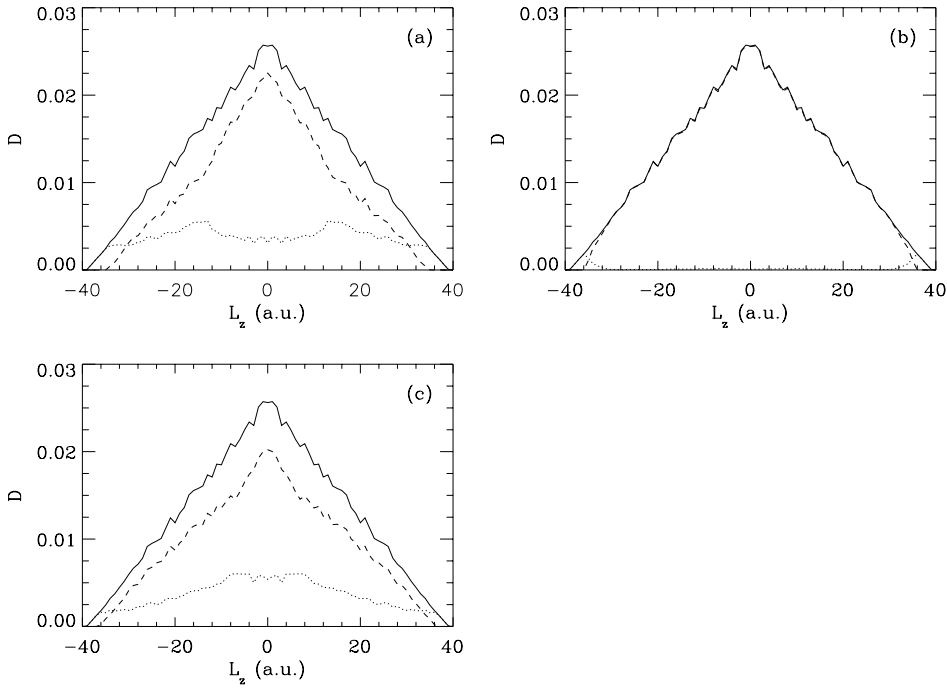


Figure 3. The distribution, D , of the z -component of the angular momentum. The initial distribution is the solid curve, the distribution for the population that survived 150 microwave periods is the dotted curve, and the distribution for the population that ionized is the dashed curve. The microwave field parameters are the same as in figure 1. (a) Static field below resonance (41 V cm^{-1}). (b) Static field on resonance (49 V cm^{-1}). (c) Static field above resonance (52 V cm^{-1}). Note that the small $|L_z|$ population has the highest probability for ionization; the population with the largest $|L_z|$ (>35) does not ionize, even at resonance.

For $17\tau \lesssim t \lesssim 35\tau$ the ionization continues at the same pace at the resonance dip, that is, the survival curve has the same slope as for $t \lesssim 17\tau$. It is during this time that the static field being at resonance plays a significant role in the process. Finally, for $t \gtrsim 35\tau$, even the ionization at the resonance shuts down.

The classical simulations in figure 3 show the original distribution of the z -projection of the angular momentum (solid curve) and the distributions for the atoms that ionized (dashed curve) and survived (dotted curve). The data in figure 3 are for

- (a) static field below resonance (41 V cm^{-1}),
- (b) static field at resonance (49 V cm^{-1}), and
- (c) static field above resonance (52 V cm^{-1}).

All the results shown in figure 3 confirm a statement that we made in a previously published analytical paper [17]: namely, that in parallel microwave and static fields the states with small $|L_z|$ are the easiest to ionize. Conversely, note that the largest $|L_z|$ (>35) do not ionize, even at the resonance.

5. Conclusions

The recent parallel-fields experiments of Koch *et al* [8] contain new challenges for atomic physicists and nonlinear dynamicists: the fields are too strong for standard time-dependent

perturbation theory to work and we lack reliable nonlinear-dynamical tools to analyse these multidimensional systems. Some aspects of the resulting ionization yield curves (regular oscillatory features and signatures of resonance transitions in the form of sharp dips) have already been explained using classical and semiclassical methods, leaving open the question of the origin of the very fast oscillations. In this paper, we report detailed simulations of the survival probability of hydrogen atoms in parallel static and microwave fields. In one simulation, we used classical trajectory Monte Carlo techniques with an isotropic distribution for the hydrogen atom before entering the field region. In the second simulation, we time-propagated Schrödinger's equation using a split operator method and a mask to allow ionization. The simulations agreed with each other and are in qualitative agreement with the slow variation of the experimental survival probability with the static field strength, F_s ; the disagreement between the classical and quantum simulation is mostly due to the different definition of survival used in the two simulations (the quantum simulation uses a mask to remove electron probability over a range of distances from the proton while the classical simulation removes trajectories at a specific distance). Neither the classical or quantum simulation contained the fast oscillation of the survival probability with the static field strength that is present in the experiments. Our quantum calculation rules out the possibility that this fast oscillation is due to quantum interference.

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