

Multiphoton ionization of hydrogen: A time-dependent theory

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Results of calculations using a novel numerical method for intense-field, low-order multiphoton ionization of hydrogen are reported and compared to previously published values. The technique involves the explicit numerical solution of the time-dependent Schrödinger equation using a finite-difference representation of the electronic wave function.

Multiphoton ionization (MPI) has been the subject of active theoretical¹ and experimental²⁻⁵ interest recently due to the development of high-power, short-pulse laser systems. As the intensities of these lasers increase and the pulse lengths become shorter, it is expected that traditional understanding of the ionization process based on standard perturbation treatments will break down. Peak intensities corresponding to an atomic unit in field strength have been achieved and will be exceeded. Under these conditions, the laser field must be treated on the same footing as the Coulomb forces in the atom. Also, as the pulse rise time becomes shorter, the bandwidth of the laser could become very significant in determining the ionization dynamics. We describe here a new computational method for studying MPI which includes these effects. In this Rapid Communication we present calculations of the one-, two-, and three-photon-dominant ionization rates of the hydrogen atom for laser intensities in the range of 10^{12} – 10^{14} W/cm². Chu and Cooper⁶ have performed calculations for these processes using an L^2 non-Hermitian Floquet method. Their ionization rates correspond to the inverse of the widths of the ground- (lowest) state eigenfunctions in the presence of the field. Their method is completely nonperturbative. As long as a sufficient number of Floquet blocks representing different numbers of photons absorbed is included, their results should be exact for a constant-intensity, monochromatic laser field.

In this work the ionization rates are obtained by direct numerical integration of the time-dependent Schrödinger equation. These calculations, which are also nonperturbative, include the effects of the temporal shape of the laser pulse. This method can be extended to treat multielectron atoms using a time-dependent mean-field or self-consistent-field formulation of the interelectronic interactions. Also, very-high-order ionization processes can be treated as easily as those presented here. The purpose of this note is to describe the method and to illustrate its application in a regime where another exact method is available. Future communications will present extension of the technique to ionization processes for intensities, wavelengths, and pulse widths for single- and multielectron systems for which no other method exists. In Sec. I, a brief description of the calculation is presented. The results for a few wavelengths and intensities are compared to those of Chu and Cooper in Sec. II.

I. CALCULATIONS

The time-dependent Schrödinger equation for a single-electron atom in a laser field is solved by direct numerical integration. The laser is assumed to be linearly polarized and the laser-atom interaction is treated in the dipole approximation. The field is treated classically. Therefore, the Hamiltonian is given by

$$H = -\frac{\hbar^2}{2m}\nabla^2 - \frac{e^2}{r} - eE(t)z \sin(\omega t), \quad (1)$$

where the amplitude of the field is chosen to be given by

$$E(t) = \begin{cases} E_{\max}t/t_{\max}, & t < t_{\max}, \\ E_{\max}, & t > t_{\max}, \end{cases} \quad (2)$$

where t_{\max} corresponds to the period during which the laser is being "turned on." This is typically chosen to be ten oscillations of the field. This turn-on time can be long enough that the intensity increase is adiabatic or can be very short if effects of pulse shape are of interest. During the constant-intensity period, an ionization rate can be determined. We choose the phase of the field to be $\sin(\omega t)$ so that the initial condition corresponds to the atom in its ground state (in the absence of the field). This Hamiltonian has cylindrical symmetry so that we can write the wave function as

$$\Phi(\mathbf{r}, t) = \sum_m e^{im\phi} \chi_m(\rho, z, t), \quad (3)$$

where, for initial s states, only the $m=0$ term exists. Putting this into the time-dependent Schrödinger equation results in a single partial differential equation in two spatial dimensions which determines the time evolution of the electronic wave function.

We solve this time-dependent equation for χ using an alternating-direction implicit propagation scheme for a finite-difference representation of the wave function.⁷ A two-dimensional grid in ρz space is chosen, and the kinetic energy is calculated using a three-point formula. The details of the calculation follow closely that which was done previously for charge exchange using a time-dependent Hartree-Fock method.⁸

The initial state is the ground state of the atom determined on the grid. We include a short-range imaginary

part to the potential on the outer edges of the grid to absorb the flux leaving the domain of the calculation. This absorbing boundary must be far enough from the nucleus that the escaping electron is no longer capable of absorbing or emitting photons due to the remaining Coulomb field. When some portion of the wave function is sufficiently excited to propagate to the boundary of the grid, it is absorbed. We assume this flux corresponds to that part of the wave function which is in the continuum. For the wavelengths considered here, this is a good assumption. Thus the time change of the norm of the wave function is taken to be the ionization rate. The solid curve in Fig. 1 shows the time dependence of the norm for a peak intensity of 1.75×10^{14} W/cm² and a photon energy of 0.2 a.u. Since it takes 0.5 a.u. to ionize the atom, this calculation is in the regime where the three-photon process is expected to dominate. During the first half of the integration time, the laser intensity increases linearly, then is constant over the remainder. During the constant-intensity period, the wave-function norm decreases exponentially. We find that the depletion of the wave function due to excitation during the turning-on period does not effect the steady decay rate observed during the constant-intensity time except for a brief transient period. The constant-intensity time interval is chosen to be long enough to obtain a well-defined ionization rate.

We also project the time-dependent wave function onto the initial state to determine what fraction remains in the ground state. This is shown by the dashed curve in Fig. 1. As can be seen there is a delay time between the probability leaving the ground state and being absorbed at the boundary. A simple calculation shows that this delay is comparable to the time it takes for the electron to propagate to the edge of the grid, a distance of $32a_0$ from the nucleus.

We have verified the rates reported here are relatively insensitive ($\pm 20\%$) to the grid size, to the magnitude and range of the imaginary part of the potential, to the rise time of the pulse, and to other reasonable variations of the

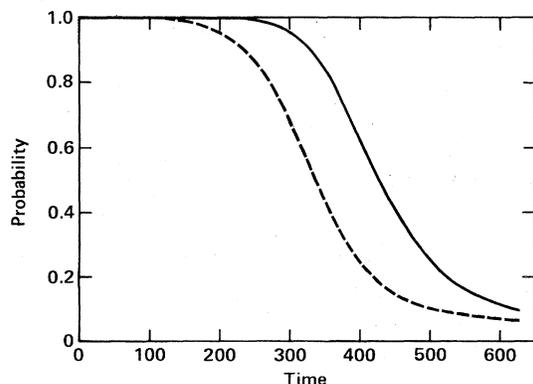


FIG. 1. Time dependence of the norm of the wave function (solid curve) and the probability remaining in the initial state (dashed curve) for a photon energy of 0.2 hartree and a maximum of intensity of 1.75×10^{14} W/cm². Time is in atomic units— 2.42×10^{-17} s. The amplitude of the field reaches its maximum at 314 a.u.

integration parameters. In these calculations we used approximately 26000 spatial grid points and 400 time steps per laser period. A 20-period calculation takes 12 min on a CRAY 1 computer.

II. RESULTS

We have calculated the ionization rate of a hydrogen atom for a selected number of laser wavelengths and intensities. The results are presented in Table I along with those calculated by Chu and Cooper⁶ using a Floquet approach. The disagreement between the two calculations is within the expected accuracy of the method described here. The agreement is excellent for single photon ionization and is least satisfactory for the third-order process. Assuming the Chu and Cooper results are converged the difference must be due to the finite grid spacing ($0.25a_0$). The error arises both from an approximate evaluation of the second derivatives in the kinetic energy operator and a sparse representation of the wave function in the immediate vicinity of the nucleus. Since our intent is to use these methods for more complicated systems where the physics of the ionization dynamics is in question, it is important that these results are reasonably accurate.

It is clear from the results of Chu and Cooper that in this intensity regime the two- and three-photon ionization rates do not obey the power laws predicted by lowest-order perturbation theory. For the frequencies studied the effective order of the process falls below the perturbation values at the higher intensities. This apparent saturation effect is reproduced in our calculations also as shown by the results for $\hbar\omega = 0.2$ a.u. As the intensity decreases both calculations should converge to the perturbation-theory rates. However, if the intensity at which this saturation effect appears is so low that the ionization rate is lower than 10^{10} s⁻¹, our time-dependent method is too expensive to use.

These calculations illustrate a new method for studying the dynamics of multiphoton ionization. The direct solution of the time-dependent Schrödinger equation can be used for very-high-order, nonperturbative calculations of ionization with pulses of arbitrary time dependence. Future communications will deal with these effects. Since the method must follow the time evolution of the electronic wave function it is not appropriate for weak field processes, for which standard perturbative methods (which are much less demanding computationally), have been shown to be accurate.

TABLE I. Ionization rates (s⁻¹) for hydrogen.

| $\hbar\omega$ (a.u.) | I (W/cm ²) | R (Ref. 6) | R (present) |
|----------------------|--------------------------|-----------------------|----------------------|
| 0.55 | 7.00×10^{12} | 1.43×10^{13} | 1.4×10^{13} |
| 0.28 | 7.00×10^{12} | 3.73×10^{11} | 3.3×10^{11} |
| 0.28 | 4.38×10^{13} | 1.33×10^{13} | 1.2×10^{13} |
| 0.20 | 4.38×10^{13} | 3.86×10^{12} | 2.8×10^{12} |
| 0.20 | 1.75×10^{14} | 2.89×10^{14} | 4.0×10^{14} |
| 0.20 | 3.94×10^{14} | 5.64×10^{14} | 7.0×10^{14} |

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