Multiphoton Ionization of H^- and He in Intense Laser Fields

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The recently proposed R-matrix-Floquet theory of multiphoton processes has been used to calculate multiphoton ionization rates for the two-electron systems H^- and He in intense laser fields. The theory is nonperturbative and includes electron-electron correlations. Results are presented for total and partial multiphoton ionization rates and novel nonperturbative correlation effects are discussed.

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The study of multiphoton ionization of atomic systems by intense laser fields has attracted considerable attention in recent years [1]. In the case of atomic hydrogen there have been many theoretical studies both within the Floquet framework [2] and also by solving the time-dependent Schrödinger equation numerically [3]. However, most theoretical work for atoms and ions containing more than one electron has relied on the use of perturbation theory [4] or the time-dependent Hartree-Fock approximation [5] which includes only a restricted class of electron correlations. Recently, the present authors [6,7] have introduced a new R-matrix-Floquet theory of multiphoton processes which is nonperturbative, enabling it to be applied for intense fields, and also allows electron-electron correlation effects to be included for an arbitrary atom or ion. The associated computer programs have recently been shown to give accurate results for atomic hydrogen [8]. In this Letter the first multiphoton ionization rates for the two electron systems H^- and He are presented which show novel nonperturbative correlation effects. The results also indicate that the method is capable of yielding accurate total and partial multiphoton ionization rates for any atomic system.

The R-matrix-Floquet theory of multiphoton processes starts from the time-dependent Schrödinger equation describing the interaction of the laser field with a general $(N+1)$-electron atom or ion, which in atomic units reads

$$i\frac{\partial}{\partial t}\Psi(\mathbf{X}_{N+1},t)=\left[H_{N+1}+\frac{1}{c}\mathbf{A}(t)\cdot\mathbf{P}_{N+1}+\frac{N+1}{2c^2}\mathbf{A}^2(t)\right]\Psi(\mathbf{X}_{N+1},t)\,. \tag{1}$$

Here $\mathbf{X}_{N+1}\equiv\{\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_{N+1}\}=\{\mathbf{X}_N,\mathbf{x}_{N+1}\}$ is the ensemble of space and spin coordinates of the $N+1$ electrons, H_{N+1} is the field-free nonrelativistic Hamiltonian and the vector potential is given by

$$\mathbf{A}(t)=\hat{\epsilon}A_0\sin\omega t\,, \tag{2}$$

which assumes that the laser field is monomode, monochromatic, linearly polarized, and spatially homogeneous. Finally $\mathbf{P}_{N+1}$ is the total electron momentum operator.

In accordance with the R-matrix method [9], configuration space is divided into an internal and an external region, in each of which the most appropriate form of the laser-atom interaction is used. The internal region is defined by the condition that the radial coordinates r_i of all $N+1$ electrons satisfy

$$r_i\le a,\quad i=1,2,\ldots,N+1\,, \tag{3}$$

where the sphere of radius a is chosen to just envelop the charge distribution of the target atom (ion) states retained in the calculation [see Eq. (5) below]. In this region electron exchange and correlation effects involving all $N+1$ electrons are important. Also the dipole-length gauge is used to describe the interaction of the electrons with the laser field.

The external region is defined so that one of the $N+1$ electrons lies outside the sphere of radius a and the remaining N "target" electrons are confined within this sphere. This outer electron corresponds physically to the ejected electron in multiphoton ionization. Since the outer electron and the remaining N electrons occupy different regions of space, electron exchange between them is negligible. Further, in the external region, the dipole-velocity gauge is used to describe the interaction of the outer electron with the laser field, necessitating a gauge transformation on the boundary of the two regions.

In order to solve the Schrödinger equation in both regions we use the Floquet-Fourier expansion of the wave function

$$\Psi(\mathbf{X}_{N+1},t)=e^{-iEt}\sum_{n=-\infty}^{\infty}e^{-in\omega t}\psi_n(\mathbf{X}_{N+1})\,. \tag{4}$$

In the internal region $\psi_n(\mathbf{X}_{N+1})$ is expanded in the R-matrix basis

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$$\psi_{kn}(\mathbf{X}_{N+1}) = \mathcal{A}\sum_{ij}\phi_i(\mathbf{X}_N)u_j(\mathbf{x}_{N+1})a_{ijkn} + \sum_j \chi_j(\mathbf{X}_{N+1})b_{jkn}\,, \quad (5)$$

where $\mathcal{A}$ is the antisymmetrization operator, the ϕ_i are a set of target eigenstates and pseudostates, u_j are basis functions representing the ejected electron which are nonzero in the boundary of the internal region, and the χ_j are quadratically integrable correlation functions, formed from the target orbital basis, which vanish by the boundary. The coefficients a_{ijkn} and b_{jkn} are obtained by constructing and diagonalizing the Floquet Hamiltonian using a modified version of the R-matrix computer programs developed for field-free electron-atom collisions and single photon ionization processes [9]. In the external region a close coupling expansion is adopted including the same target basis states as in Eq. (5) and the resulting coupled differential equations describing the motion of the ejected electron are solved out to a large radius a' where an asymptotic expansion is applied [7]. The wave functions on the boundary of these two regions are matched through the R matrix. For multiphoton ionization, outgoing wave "Siegert" boundary conditions are then imposed using an iterative technique in the complex energy plane, where the imaginary part of the corresponding energy eigenvalue gives $-\frac{1}{2}$ times the total multiphoton ionization rate. Partial rates and angular distributions are obtained by resolving the corresponding eigenvector into its channel components. Finally we note that in order to obtain converged results at the highest field intensities considered in this Letter we had to retain eight terms in the Floquet-Fourier expansion (4). This meant that after the usual partial wave decomposition of the R-matrix equation (5) we had to solve a problem with up to 90 coupled channels.

We first present our results for multiphoton detachment of H^-. We carried out calculations using two approximations. In the first, the $1s$ atomic hydrogen ground state together with $\overline{2s}$ and $\overline{2p}$ pseudostates were retained in Eq. (5). The $\overline{2p}$ pseudostate is that proposed by Damburg and Karule [10] and gives the full dipole polarizability of the $1s$ ground state, while the $\overline{2s}$ pseudostate allows for additional radial correlation effects. The lowest $^1S^e$ R-matrix pole is then adjusted so that this model gives the exact H^- attachment energy of 0.02775 a.u. In the second approximation, the $1s$, $2s$, and $2p$ target eigenstates were retained in expansion (5). This enables multiphoton detachment rates in the neighborhood of the doubly excited resonances below the $n=2$ threshold [11] to be studied.

The accuracy of this $1s$-$\overline{2s}$-$\overline{2p}$ approximation is assessed by comparing in Fig. 1 the two-photon detachment rates in the perturbative limit, corresponding to a field of intensity of 10^9 W cm^{-2}, with perturbation theory calculations of Liu, Gao, and Starace [12] and Proulx and Shakeshaft [13]. The good agreement which we obtain indicates that electron-electron correlation effects are well represented by the $1s$-$\overline{2s}$-$\overline{2p}$ model in this low energy region. In Fig. 2 two-photon detachment rates calculated in the $1s$-$2s$-$2p$ approximation near the $n=2$ threshold are shown for a field of intensity 10^{11} W cm^{-2}. A series of resonances converging to this threshold, with $S=0$ (singlet) symmetry and even parity allowed by spin and parity conservation, are clearly seen. The lowest energy resonance occurring near 0.352 a.u. has $^1S^e$ symmetry, the second resonance occurring near 0.373 a.u. has $^1D^e$ symmetry and the third very narrow resonance occurring near 0.374 a.u. has $^1S^e$ symmetry. These results are in accord with the field-free resonance positions and widths calculated in the $1s$-$2s$-$2p$ close coupling approximation [11]. Higher resonances in these series can be obtained by our method by carrying out the calculations using a

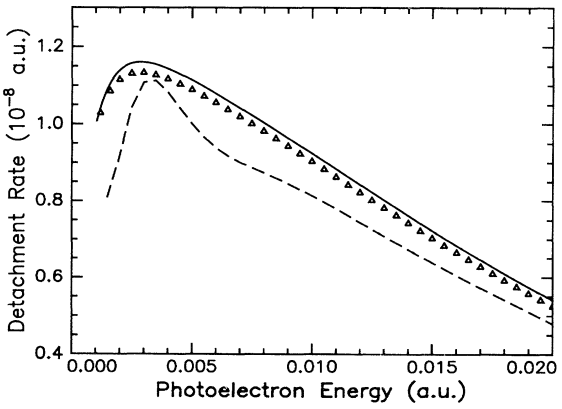

FIG. 1. Two-photon detachment rate of H^- vs photoelectron energy close to threshold at an intensity of 10^9 W cm^{-2}. Solid line, present $1s$-$\overline{2s}$-$\overline{2p}$ calculations; Δ, Liu, Gao, and Starace [12]; dashed line, Proulx and Shakeshaft [13].

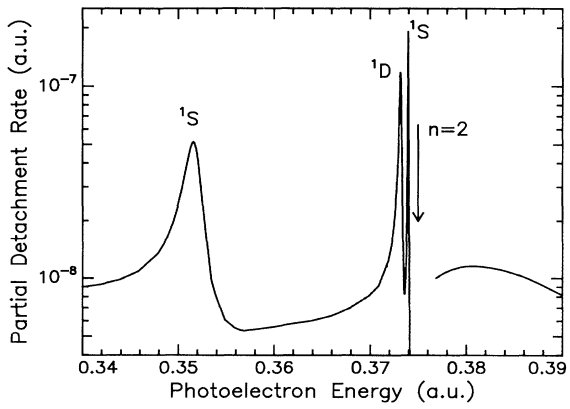

FIG. 2. Two-photon detachment rate of H^- vs photoelectron energy near the $n=2$ threshold at an intensity of 10^{11} W cm^{-2}. Present $1s$-$2s$-$2p$ calculations showing resonance structure. The position of the $n=2$ threshold is denoted by an arrow.

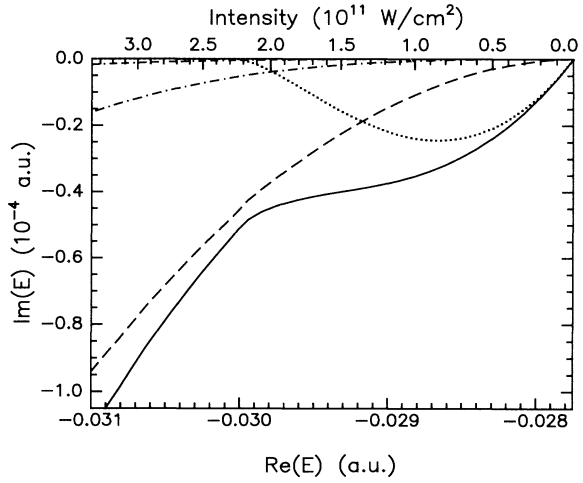

FIG. 3. Trajectory of the H^- state in the complex energy plane in the $1s$-$\overline{2s}$-$\overline{2p}$ approximation as a function of field intensity for $\omega=0.03$ a.u. Solid line, total absorption half-width; dotted line, one-photon absorption half-width; dashed line, two-photon absorption half-width; dashed-dotted line, three-photon absorption half-width; short dashed-dotted line, four-photon absorption half-width. The corresponding intensity is given on the top axis.

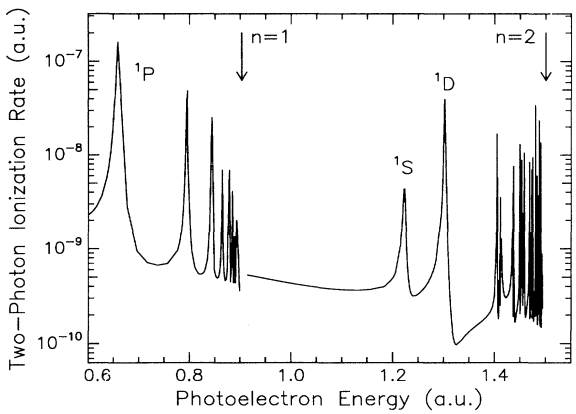

FIG. 4. Two-photon ionization rate of He vs photoelectron energy at an intensity of 10^{12} W cm^{-2}. The positions of the $n=1$ and $n=2$ thresholds are denoted by the arrows.

finer energy mesh.

At higher intensities perturbation theory is no longer applicable. An example of this regime is shown in Fig. 3 where the trajectory of the H^- state in the complex energy plane, calculated in the $1s$-$\overline{2s}$-$\overline{2p}$ approximation, is plotted as a function of field intensity for $\omega=0.03$ a.u. The threshold energy is fixed at $\omega=0$ and the shift of the ground state energy is monotonic in intensity although not quite linear. We note that by viewing the figure upside-down we can interpret it as giving multiphoton ionization rates versus intensity. In the zero-field limit the corresponding eigenvalue lies on the real energy axis with $E=-0.02775$ a.u. As the laser field is switched on the eigenvalue moves into the lower half complex energy plane since H^- can then ionize by absorption of one photon. For larger field strengths more than one photon can be absorbed with appreciable probability and the corresponding partial widths, which are also plotted in this figure, become nonzero. Finally, for field strengths greater than 2.3×10^{11} W cm^{-2} the real part of the energy eigenvalue has shifted below -0.03 a.u. Ionization by one photon absorption is then no longer energetically possible and the total width is due to absorption of two or more photons. The resultant trajectory in the complex energy plane experiences a sharp bend due to the channel closing at this field strength which is clearly nonperturbative in character and should have observable consequences. We also note the fact that, even though ω is larger than the weak-field binding energy, the shift is negative due to electron-electron correlation effects.

Turning now to multiphoton ionization of He, the calculations in this case are all carried out retaining the $1s$, $2s$, and $2p$ He^+ target eigenstates in Eq. (5). Figure 4 shows the ionization rate into the two-photon channel, versus photoelectron energy, for He in a field of intensity 10^{12} W cm^{-2}. At this intensity, we expect perturbation theory to be valid and indeed we obtain excellent agreement with the perturbative results of Proulx and Shakeshaft [14]. The Rydberg series of peaks visible below the $n=1$ threshold are all $^1P^o$ one-photon bound state resonances. In lowest order perturbation theory these resonances would all have infinite height (and thus zero width). That they do not is already a signature of the nonperturbative character of our theory. The series of resonances below the $n=2$ threshold are composed of $^1S^e$ and $^1D^e$ autoionizing state resonances in the final (two-photon) channel.

In order to exhibit nonperturbative effects we show in Fig. 5 the total ionization rate over the energy interval around the two-photon resonance between the ground state and the lowest $^1D^e$ resonance. At low intensities the total rate is dominated by one-photon absorption and hence increases linearly with intensity in the absence of resonances. We have therefore scaled the rates as indicated in the figure caption. At higher intensities we observe the emergence of a pronounced resonance structure due to the coupling with the $^1D^e$ autoionizing state which, in the absence of the field, has a natural width of 0.00269 a.u. in agreement with earlier work [11].

In conclusion multiphoton ionization rates, obtained using the R-matrix-Floquet theory, have been presented for two-electron systems, which are both nonperturbative and also include electron-electron correlation effects. The computer programs which have been developed can be used to obtain multiphoton ionization rates and laser assisted electron collision cross sections for any atomic system. In the future it is intended to apply these programs

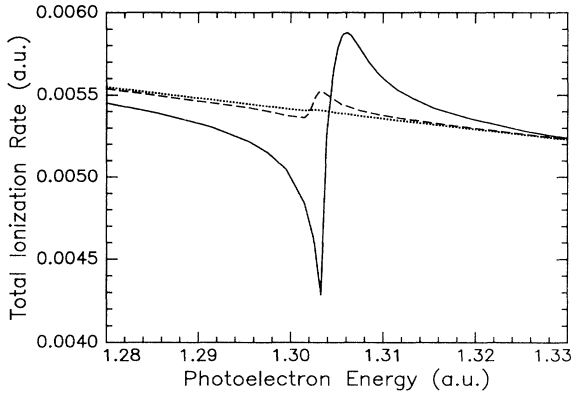

FIG. 5. Total ionization rate of He vs photoelectron energy for two-photon absorption. Dotted line: intensity 2×10^{12} $W cm^{-2}$, rate×100. Dashed line: intensity 2×10^{13} $W cm^{-2}$, rate×10. Solid line: intensity 2×10^{14} $W cm^{-2}$.

to study the heavier inert gas targets where there is currently considerable experimental interest.

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