Theory of Intense-Field Ionization and Above-Threshold Ionization of Helium through Multiple Resonances

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We present a quantitative theory of ionization and above-threshold ionization of He by short (subpicosecond), intense laser pulses. The calculations include the effect of bound states shifting through resonance during the pulse as well as the switching of the process from six to seven photons due to the shift of the ionization threshold. The results are in good agreement with and add further insight into recent experimental data.

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Two recent experiments by Perry and collaborators[1,2] have provided valuable pilot studies of the behavior of complex atomic structure under strong, short, near-UV pulses. The importance of such experiments lies in the opportunity they offer for the understanding of the interplay between strong-field phenomena and the electron-electron interactions underlying the structure of all atoms except hydrogen. The theoretical interpretation of such experiments demands the proper treatment of the atom-field interaction dynamics as well as the intra-atomic multielectron dynamics, each of which represents a challenging problem in itself.

An approach[3] based on a density-matrix theory of the laser interaction dynamics combined with atomic multichannel-quantum-defect-theory (MQDT) parameters[4] has produced very good agreement with the experimental results[1] on Kr. Some remaining rather minor discrepancies in detail may be due to inadequacies of MQDT as employed in this problem, sensitivity to the spatial distribution of the laser intensity or, conceivably, but not likely in that case, the onset of nonperturbative effects.

The newest experiment by Perry, Szoke, and Kulander[2] in He provides an excellent context for the evaluation of certain basic questions, such as the following: How far can the density-matrix theory with perturbatively calculated parameters go? How far can a single-electron model of He be stretched in the description of this interaction? The attractiveness of this problem stems from the possibility it offers for the evaluation of departures of the atomic parameters from the single-electron approximation as discussed below. The results obtained here can thus serve as benchmarks for approaches involving nonperturbative techniques[5-7] which, being heavily numerical, do not always allow intuitive explanations of the underlying physics. Compared to the work in Kr,[1] this problem provides a glimpse at the behavior at significantly higher intensity with the simultaneous involvement of many states, the presence of above-threshold ionization (ATI), and a Stark shift of the ionization potential sufficiently large to change the order of the process from six to seven photons during the pulse. In short, we have most of the ingredients of strong-field atomic interactions in the simplest two-electron system.

Formulation and theoretical model.— Let $|0\rangle$ be the atomic ground state and $|j\rangle, |j'\rangle, \ldots$ excited states which are or are apt to become resonant with some m-photon transition during the pulse. At zero (or relatively weak) field and photon energy 4.40 eV ($\lambda = 281.5$ nm), a minimum of six photons is required for the ionization of helium with an ionization potential (IP) of 24.58 eV. The energy of five photons (5ω) is 0.78 eV above the energy 21.22 eV of the unperturbed $1s2p(^1P_1)$ state. At weak field, the gross features of the six-photon ionization could be described in terms of a two-state density matrix, containing $1s^2$ and $1s2p$. At the intensities employed in the experiment, however, other states will come into six-photon resonance as the IP shifts above the six-photon level. Thus the number of states that play a dominant role in the overall process changes during the pulse. The range of peak intensities we are examining here reaches up to 5.6×10^{14} W/cm^2.

Let us consider the values of some relevant ac Stark shifts at the frequency of the experiment. The ground state has a small negative (downward) shift of -0.04 eV per 10^{14} W/cm^2. The Rydberg states and the IP have positive (upward) shifts practically equal to the ponderomotive shift S_p, which in atomic units is given by $I/4\omega^2$, where the laser intensity I and the frequency ω must also be inserted in atomic units. It corresponds to ~0.74 eV per 10^{14} W/cm^2, which, if compared with the difference $6\omega - \mathrm{IP}$, shows that, at the intensity $I_6 \simeq 2.4\times10^{14}$ W/cm^2 and above, six-photon ionization is not possible. We have the first so-called channel closing. Above I_6, the process is best described as six-photon-resonant, seven-photon ionization. At the maximum peak intensity

employed by Perry, Szoke, and Kulander[2] (4.4×10^{14} W/cm^2), the ponderomotive shift is ~3.25 eV. Given that all excited states of the form $1snl(^1L_l)$, with the exception of $1s2p$ (see Table I), shift by amounts very close to S_p, we find that even at the above highest peak intensity the energy of 6ω is still above that of the $1s3d(^1D_2)$ but below the rest of the excited states. We expect therefore that up to that intensity, only even-parity states of the form $1sns$, $1snd$, etc., $n\geq4$, may give rise to significant resonant enhancement. For n too large, the states are spaced too closely to produce non-negligible enhancement. It is now the relative magnitude of atomic parameters for the various states that will help us sort out those that shape the observed behavior. The important parameters and states are the following: the five-photon Rabi frequency to the $1s2p$; the six-photon Rabi frequencies to the states $1s3d$, $1s4s$, $1s4d$, $1s5s$, $1s5d$; and the one-photon and two-photon (involving one ATI) ionization cross sections of each of these states, and their ac Stark shifts. We have listed a set of values for those parameters in Table I. Note that the shift of $2p$ is much larger than S_p (in this wavelength range) while that of $3d$ is smaller by about 10%. The rest of the shifts are essentially equal to S_p. Deferring for the moment discussion of the calculation of these parameters, we note that together with the ground state they constitute a set of seven states which we chose (after evaluation of their positions during the pulse) as the basis for the analysis of the interaction dynamics.

The calculations involved the solution of a 7×7 system of density-matrix equations. We present here a special case of these equations in order to discuss certain aspects of the method. With the notation for the ground and excited states established above, a system of 3×3 equations is written as

$$\frac{d}{dt}\sigma_{00}=\mathrm{Im}(\Omega_{01}\sigma_{10}+\Omega_{02}\sigma_{20}), \tag{1a}$$

$$\frac{d}{dt}\sigma_{jj}=-\Gamma_j\sigma_{jj}-\mathrm{Im}(\Omega_{0j}\sigma_{j0}),\quad j=1,2, \tag{1b}$$

$$\frac{d}{dt}\sigma_{j0}=\frac{i}{2}\Omega_{j0}(\sigma_{jj}-\sigma_{00})+\frac{i}{2}(\Omega_{j'0}\sigma_{jj'})+\frac{i}{2}(2\Delta_j+i\Gamma_j)\sigma_{j0},\quad j,j'=1,2\ (j\neq j'), \tag{1c}$$

$$\frac{d}{dt}\sigma_{12}=\frac{i}{2}(\Omega_{02}\sigma_{10}-\Omega_{10}\sigma_{02})-\frac{i}{2}[2\Delta_{12}-i(\Gamma_1+\Gamma_2)]\sigma_{12}. \tag{1d}$$

In this sample system of equations, $|j\rangle,|j'\rangle$ denote Rydberg states that can come into six-photon resonance with the ground state $|0\rangle$, with Ω_{0j} being the respective six-photon Rabi frequencies, and Δ_j the dynamic detuning from resonance defined by

$$\Delta_j=6\omega-(E_j+S_j)+(E_0+S_0), \tag{2}$$

where S_j,S_0 denote the respective Stark shifts. The ionization width Γ_j for each state is given by the sum of its one- and two-photon ionization rates which are obtained from the respective cross sections as $\sigma_j^{(1)}I$ and $\sigma_j^{(2)}I^2$, where the intensity (I) must be inserted in units of photon flux.

Using the shifts of Table I, one easily finds that the state $1s2p$ will come into five-photon resonance at an intensity of 0.7×10^{13} W/cm^2, well below I_6. By the time I_6 is reached, one can again easily deduce that $1s2p$ has shifted by ~2.1 eV above the five-photon resonance. This means that below I_6, only one excited state comes into resonance. The dynamics are then governed by a 2×2 density matrix. Above I_6, we have the 7×7 system, as the six-photon resonances begin playing a role. In the solution of the differential equations, we begin with the 2×2 and only above I_6 does the full 7×7 system come into the calculation. The reverse happens during the fall

TABLE I. Atomic parameters for $N+1$ and $N+2$ (ATI) resonant ionization of He calculated through single-channel quantum-defect theory. Within parenthesis are numbers calculated for $\hbar\omega=0.162$ a.u. and correlation effects included in the ground state (Ref. 10) and static exchange approximation for excited states. The relation between the multiphoton matrix element $r_{0i}^{(N)}$ and the Rabi frequency $\Omega_{0i}^{(N)}$ is (in a.u.) $\Omega_{0i}^{(N)}=2r_{0i}^{(N)}(I/I_0)^{N/2}$. I is the intensity in W/cm^2 and $I_0=14.038\times10^{16}$ W/cm^2.

State	E (eV)	$N(\hbar\omega)$	$r_{0i}^{(N)}$ (a.u.)	S_i [a]	$\sigma_i^{(1)}$ [b]	$\sigma_i^{(2)}$ [c]
$1s^2$				−0.04(0.028)		
$2p$	21.22	5	2158(3338)	1.16(1.1)	7.65(6.6)	1197.6(315.0)
$3d$	23.07	6	−14662	0.65(0.66)	0.234(0.42)	13.23
$4s$	23.67	6	−5948	0.73	1.119	340.3
$4d$	23.74	6	−9228	0.70	0.109	7.12
$5s$	24.01	6	−4424	0.74	0.579	179.0
$5d$	24.04	6	−5854	0.74	0.0579	4.00

[a]ac Stark shift of state i [eV/(10^{14} W/cm^2)].
[b]Single-photon ionization cross section of state i [10^{-18} cm^2].
[c]Two-photon ionization cross section of state i [10^{-53} cm^{4}s].

of the pulse.

A realistic temporal profile of the form $f(t)=I_0/\cosh^2(1.76t/\tau)$ was employed in our calculations [with the full-width-half-maximum value (τ) taken equal to 700 fs], as well as proper spatial averaging in the transverse direction using the distribution given by Perry and Landen.[1]

Atomic parameters.—The calculation of the atomic parameters listed in Table I was of particular concern in this work because it required an assessment of the importance of correlation. It is useful to note first that even the energy of the ATI peak observed in the experiment was well below the first doubly excited state of He which lies about 32 eV above the first ionization threshold. As a consequence of the large energy separation of these states from the first threshold, the quantum defects of the singly excited states do not show significant anomalies (perturbations). One might thus expect that single-channel quantum-defect theory (SQDT) would be adequate for this study. It is, however, known that SQDT does not represent the wave function well for small r, which could be critical in this case since the ground state of He is known to involve significant correlation.

First, we obtained a set of values for the parameters using SQDT (see Table I). In this part of the calculation, we employed techniques[4,8,9] and programs that we have developed and tested in a number of atoms. In a parallel work,[10,11] we calculated some of these parameters using a configuration-interaction (CI) representation for the ground state (which gives the value -2.9024 a.u. for its binding energy) combined with a discretized basis to represent the spectrum of excited states in the static exchange approximation (SEA). This basis—which includes the single-electron continuum—has also been employed in performing the summations over complete sets of intermediate states in the calculation of the multiphoton Rabi frequencies, the Stark shifts, and the two-photon ATI cross sections from the excited states. We have discussed in a recent paper[10] the method we employ in the calculation of ATI with a discretized representation of the continuum. Additional aspects requiring attention in this context have to do with the gauge, since multiphoton matrix elements of multielectron atoms are sensitive to different parts of the spectrum in different gauges (for a discussion of this point, see Bachau, Lambropoulos, and Tang[10]).

To summarize briefly, we have found SQDT to yield Rabi frequencies somewhat smaller (by about a factor of 2) than those that include correlation. The photoionization cross sections and ac Stark shifts, on the other hand, are given fairly accurately by SQDT. The five-photon Rabi frequency of the $1s2p$ appears to be particularly sensitive to the presence of correlation in the ground state. From exhaustive CI calculations in connection with nonperturbative studies that will be published in the near future, we have learned that as long as ATI does not reach the doubly excited states, He is represented fairly well by a frozen-core (FC) approximation for the excited states (with one electron frozen in the $1s$ state of He^+). SQDT gives a reasonable but not as good overall approximation, with discrepancies of no more than a factor of 2. It does, on the other hand, provide a versatile method for the calculation of a consistent set of parameters. Since the decisive parameters in the resonance structure of the experiment were the ionization widths and Stark shifts which are given satisfactorily by SQDT, we have chosen to present here the main analysis in terms of SQDT parameters.

Results and discussion.—The main body of our results pertaining to the experiment is encapsulated in Fig. 1 showing the photoelectron spectrum for three different peak intensities up to and slightly above the highest value reported in the experiment. At the lower intensities, the positions of the two peaks and their relative heights are in good agreement with the experiment. With increasing intensity, the photoelectron peaks shift only slightly towards lower energy, which confirms their relation to a resonant state. The values of our calculated shifts are also compatible with the calculated positions of states $1s2p$ and $1s3d$ reported by Perry, Szoke, and Kulander.[2] The relative values of our Rabi frequencies and ionization rates reproduce the relative heights of the photoelectron peaks quite well which lends confidence in the values of our one- and two-photon ionization cross sections.

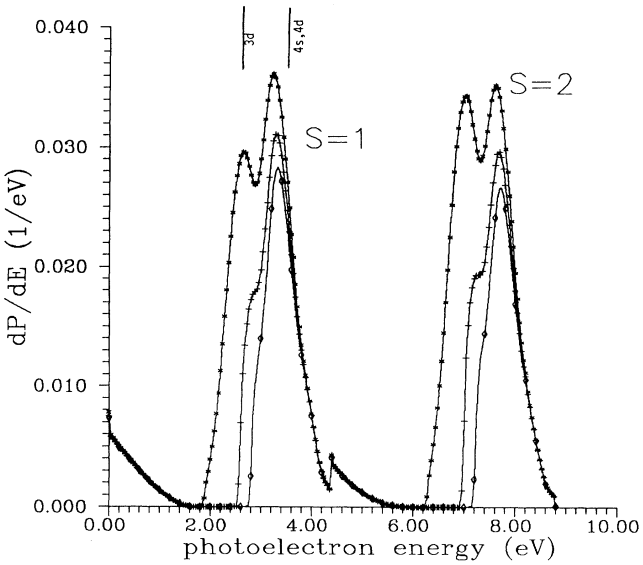

FIG. 1. Photoelectron spectrum for seven-photon ionization of helium calculated for $\lambda=281.5$ nm, $\tau=700$ fs, and three different intensities. Curves with $\diamond$, $+$, and $\times$ denote $I_0=4.4\times10^{14}$ W/cm^2, $I_0=4.7\times10^{14}$ W/cm^2, and $I_0=5.6\times10^{14}$ W/cm^2, respectively. The photoelectron energies corresponding to resonant excitation of the levels $3d,4s,4d$ during the pulse are indicated above the $S=1$ peak.

At the higher intensities of the experiment, a partially resolved feature at photoelectron kinetic energy of 3 eV with a wing extending down to 2 eV has been reported,[2] and attributed to resonance enhancement due to the state $1s3d$. At those intensities, our calculations do not produce that feature, but only one peak at about 3.8 eV as the intensity is varied from 3.1×10^{14} to 4.7×10^{14} W/cm^2. It can be traced to the six-photon resonant enhancement via the $1s4s$ and $1s4d$ states. The $1s3d$ does not produce significant enhancement (peak) until peak intensities above 5×10^{14} W/cm^2, and it is at about 5.6×10^{14} W/cm^2 that two well-separated satellite peaks appear, one corresponding to the $3d$ and the other to the $4s$ and $4d$. This pair of satellites is also mirrored in the ATI peak as they should be. We are thus led to a slight reinterpretation of the data of Ref. 2. We can reproduce all features of their photoelectron spectrum under the condition that their maximum peak intensity was $\sim25\%$ higher than reported. This would also be more consistent with the calculated shifts reported in Ref. 2. We are confident that we have accurate values for the shifts which show that the $1s3d$ cannot come into resonance below 4.8×10^{14} W/cm^2 at a wavelength of 281.5 nm. The absence of satellite structure in the experimental data for the ATI peak is perfectly compatible with the inevitable decrease of the experimental resolution as the photoelectron kinetic energy increases. As the authors of Ref. 2 have noted, that structure was revealed when they took data (not shown in their Fig. 2) with "higher-energy resolution." Finally, it should be noted that the small cusp to the right of the first main peak of our Fig. 1 (at 4.4 eV) corresponds to the region of energy where ionization switches from six to seven photons.

We have not attempted to adjust parameters so as to better match our relative heights of peaks and satellites with those of the experiment. It can be easily accomplished, as these quantities are quite sensitive to the values of ionization widths and Rabi frequencies (see Ref. 12). For such an adjustment to be meaningful, we would need to fold in the experimental resolution taking into account experimental error margins. Such an exercise is, in principle, useful in that it can provide error bars for the theoretical parameters, but it is beyond the scope of this paper. What is perhaps meaningful is to compare an effective rate of ionization obtained with our numbers to an equivalent result given in Ref. 2. Thus at 2×10^{14} W/cm^2 and 281.5 nm, we find a rate of 4×10^{10} s^{-1} as compared to about 6×10^{10} s^{-1} given in Fig. 3 of Ref. 2. Note, however, that our detailed calculations do not involve a rate but the amount of ionization as a function of time.

In conclusion, we have provided a quantitative interpretation of the experiment by Perry, Szoke, and Kulander,[2] have demonstrated that even at $\sim5\times10^{14}$ W/cm^2 the behavior can be understood in terms of the time-dependent density-matrix theory, and have found that in this wavelength range most of the influence of correlation comes through the ground state. This will not necessarily be the case at much higher frequencies or in other two-electron atoms such as the alkaline earths where the doubly excited states begin much closer to and in some cases below the first ionization threshold. Correlation in excited states would then also play a role.

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