Rydberg states in the strong field ionization of hydrogen by 800, 1200 and 1600 nm lasers

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Abstract

We study the population of Rydberg excited states in the strong field interaction of atomic hydrogen with 800, 1200 and 1600 nm laser pulses. The total excitation probability displays strong out-of-phase modulation with respect to the weak modulation in the total ionization probability as the laser intensity is increased. The results are explained in terms of channel closing, to demonstrate multiphoton ionization features in the strong tunnel ionization regime. We also explain the stability of high Rydberg states in strong laser fields in contrast to other previous ionization stabilization models.

Keywords: tunnel ionization, multiphoton ionization, Rydberg states

(Some figures may appear in colour only in the online journal)

1. Introduction

Before the invention of lasers in the early 1960 s, the description of the interaction of light with atoms and molecules was simple—it was given by the first-order perturbation theory. With lasers, for many years, the nonlinear physics was very much limited to the study of multi-photon excitation or ionization processes. Thus the seminal tunneling ionization paper of Keldysh [1] in 1965, which offers an alternative route to strong field ionization, did not attract much attention until the 1980s. With the emergence of short intense Ti–sapphire lasers operating near the 800 nm wavelength and the subsequent mid-infrared wavelength lasers, today tunneling ionization is the starting point for the understanding of almost all aspects of the strong field interaction of infrared light with atoms and molecules.

Tunnel ionization is a well-known phenomenon in quantum physics. It is used to describe the ionization of an atom in a static electric field as well as the α-decay of a nucleus. A laser is not a static electric field, but for low-frequency driving lasers the typical time scale of the motion of a valence electron in atoms or molecules is much shorter than the optical period of the laser, such that the static tunnel ionization model serves as a very good first-order approximation. Since tunneling occurs when an electron is near the outer edge of an atom, the static tunnel ionization model can be easily approximated by a one-electron model, even for a multielectron atom. The compact expression derived by Ammosov, Delone and Krainov [2]—called ADK theory—for describing the tunneling ionization of an atom, has helped to advance the understanding of the strong field physics of atoms. In the last decade, the extension of the ADK model to molecules—called MO-ADK theory by Tong *et al* in 2002 [3–5]—has further helped the advance of our understanding of the strong field ionization of molecules.

Tunneling ionization is also essential for the description of rescattering phenomena such as high-order harmonic generation (HHG), high-energy above-threshold ionization

(HATI) photoelectron spectra and nonsequential double ionization (NSDI). These phenomena are qualitatively described by the three-step model where the first step is tunnel ionization and the second step is acceleration of the released electron by the oscillating laser's electric field that would drive the electron back to recollide with the parent ion. HHG, HATI and NDSI are subsequent electron–ion collision events, i.e., photo-recombination, electron–ion elastic scattering, and electron–ion impact ionization phenomena, respectively. According to the quantitative rescattering theory [6–9], the tunnel ionization and propagation of these electrons in the laser field generate a coherent electron wave packet. The magnitude of the complex electron wave packet depends on the tunnel ionization rates.

Despite its simplicity and wide utility, the accurate quantitative theory of tunnel ionization for typical many-electron atoms and molecules is still mostly not available. Since tunneling theory is a model for describing static field ionization, it is based on adiabatic approximation and thus there is an inherent error. Furthermore, a laser is not a static field and one expects the ionization rate be dependent on the laser's wavelength. As the intensity decreases, tunneling becomes less efficient. Ionization may proceed via the absorption of multiple photons instead. In the multiphoton ionization (MPI) regime, the ionization rate may be strongly enhanced if the excited states are in resonance with the absorption of multiple photons. Thus in spite of the wide popularity of tunnel ionization theory, one has to be wary of its applicability to actual experimental situations. In principle, the validity of the theory can be tested with experimental data, but this is rarely possible for strong field experiments. Except for very few cases, strong field experiments are carried out with a short, well-focused laser beam in a gas medium. The spatial and temporal distributions of the laser pulse are generally not fully characterized. Thus accurate comparison between theory and experiment is difficult to carry out. Neither the ionization rate nor the probability can be accurately calculated based on theory if the target is a multi-electron atom or molecule. In fact, even for the simplest atomic hydrogen, many important fine features have not been carefully examined so far. Some of these fine features are expected to be present in more complex targets as well.

In this contribution we take a close look at the strong field excitation and ionization of atomic hydrogen. For this system, with care, an accurate numerical solution of the time-dependent Schrödinger equation (TDSE) in a linearly polarized 800 nm laser pulse can be carried out. We will treat such numerical results as 'experimental' data to draw conclusions on the region of laser intensities where the tunnel ionization model is accurate. We also show similar calculations using 1200 nm and 1600 nm laser pulses. Furthermore, we will examine fine features in the calculated results that are not present in the tunnel ionization model—in particular, the population of excited states and how they can be explained in terms of the multiphoton ionization model. Before closing, we will also discuss the stability of excited states in the laser field, and we will comment on other models, such as frustrated tunnel ionization [10], ionization stabilization [11–13]
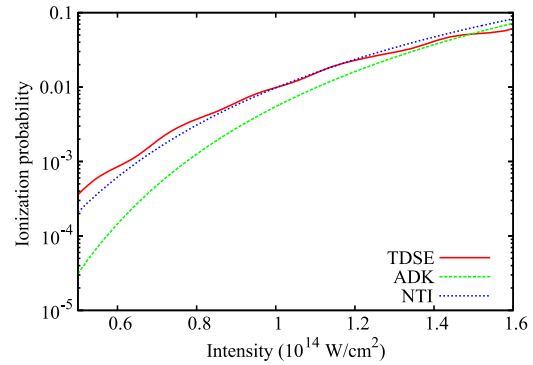

Figure 1. Total ionization probability vs intensity for atomic hydrogen in a 5 fs, 800 nm laser pulse calculated from solving the TDSE and from the ADK model. For comparison, the nonadiabatic tunneling ionization (NTI) theory of Yudin and Ivanov [17] is also presented.

and interference suppression [14, 15], that have been proposed in the past decades.

We solved the TDSE using two different methods: one using a velocity gauge and another using a length gauge, with small incremental intensity steps. Details of these calculations and convergence tests are given in a recent publication [16]. Here we focus on the analysis of the calculated results.

2. Region of validity of tunneling ionization theory vs laser intensity and wavelength

First we show the total ionization probability of atomic hydrogen for a 5 fs, 800 nm laser pulse for a peak intensity from 0.5 to 1.6×10^{14} W cm^{-2} in figure 1, calculated from the TDSE, and compare it to the result from the ADK model. In this range, the intensity increases by a factor of three, the Keldysh parameter (γ) ranges from 1.5 to 0.84, while the total ionization probability from the TDSE increases by a factor of almost 150, indicating the strong nonlinearity of the ionization process. At $\gamma = 1.0$ (an intensity of 1.1×10^{14} W cm^{-2}), which is normally taken as the dividing line of tunneling versus multiphoton ionization regimes, the ADK probability is about half of the TDSE one. As laser intensity increases, the agreement between the two calculations improves.

Above $\gamma = 1.0$ or an intensity below 1.1×10^{14} W cm^{-2}, the ADK probability drops increasingly, well below the TDSE one, reaching a factor of about ten smaller at $\gamma = 1.5$ or at an intensity of 0.5×10^{14} W cm^{-2}. Qualitatively, it is understood that tunneling ionization is no longer the main mechanism for ionization. Instead, multiphoton ionization is perceived as a more efficient means for removing the electron from the atom, with the understanding that for short pulses this is a broadband (a width of 0.36 eV for a 5 fs pulse) photon. Since it would take about eleven photons (of 1.55 eV) to ionize atomic hydrogen for a laser at 0.5×10^{14} W cm^{-2}, a direct calculation of the ionization probability based on perturbative multiphoton ionization theory is not practical. Thus there is no proof that the multiphoton ionization model alone can explain the TDSE result in this intensity region.
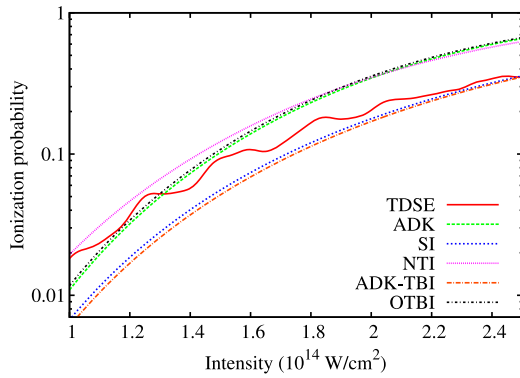
Figure 2. Total ionization probability vs intensity for atomic hydrogen in a 10 fs, 800 nm laser pulse calculated from solving the TDSE and from the ADK model are shown over the higher laser intensity region. Also shown are the model of Yudin and Ivanov (NTI) [17], the static ionization (SI) theory and the ADK–TBI model of Tong *et al* [18], and the over-the-barrier modification model (OTBI) from Krainov [19].

Figure 3. Comparing total ionization probability by 10 fs laser pulses with 800, 1200 and 1600 nm lasers, respectively, calculated from solving the TDSE. Probabilities calculated from ADK theory and static field ionization theory are found to be independent of wavelengths. As the wavelength is increased, the TDSE result approaches the static field ionization limit.

Since ionization is the first step for any strong field phenomena, it is desirable to find a means to 'improve' or 'extend' the validity of the simple ADK model to a broader intensity region. In figure 1, we also show the ionization probability calculated based on the model of Yudin and Ivanov [17]. This nonadiabatic tunnel ionization (NTI) model incorporates the nonadiabatic effect (beyond the static tunneling mechanism), and thus the probability calculated from this model indeed improves the agreement with the TDSE result in the lower intensity regime. We mention that, in all of the calculations reported in this work, the laser is taken to have a sine-square pulse shape, and the carrier-envelope phase is fixed at zero. To be specific, the ADK theory used here follows the prescription of Tong and Lin [18].

In figure 2, we examine ionization for intensities from 1.0 to 2.5×10^{14} W cm^{-2}, for γ from about 1.0 to 0.68, i.e., in the so-called tunneling region. Within this range, the ionization probability from the TDSE increases only by about a factor of ten when the intensity increases by a factor of 2.5. In this higher intensity regime, the ADK theory over-estimates the ionization probability from the TDSE, as does the theory of Yudin and Ivanov, which is based on the ADK model. The nonadiabatic effect introduced in this model is not important in this tunneling ionization regime.

The ADK theory is derived for static tunneling ionization in the perturbation limit. It fails when the electric field is large. For a simple one-electron atom, static field ionization can be calculated accurately. For practical applications, it is desirable to have a simple expression for the ADK model to calculate the ionization probability even at higher intensities. In Tong and Lin [18], a damping factor was introduced empirically into the exponential function in the ADK expression for the ionization rate. The empirical function is obtained by fitting the modified rate, called the ADK–TBI model, such that the rate agrees with the calculated static ionization rate. The results from this ADK–TBI model, as shown in figure 2, agree with the static ionization (SI) probability and both are much closer to the TDSE results at higher
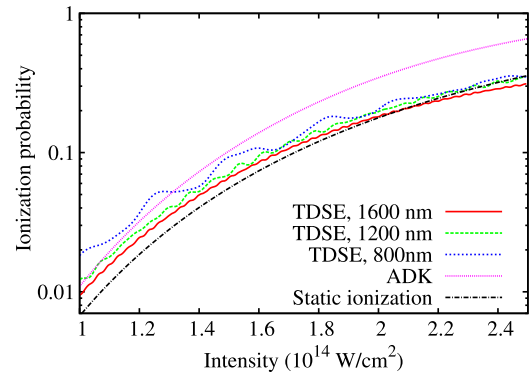
laser intensities. We comment that a modified ADK model (denoted as OTBI here) was proposed earlier by Krainov [19] for the high intensity regime. Our calculations indicate that this OTBI model gives identical results to the ADK model throughout the intensity range studied here, i.e., it fails to improve the simple ADK model.

In figure 2, upon close examination, we note that the ionization probabilities from TDSE show clear weak modulations with intensity. This modulation is not present in all the model calculations employed in the figure. Recall that all the models used are based on the static tunneling ionization theory.

To examine the wavelength dependence of the ionization probability, in figure 3, we consider the ionization of H for intensities in the $1.0 - 2.5 \times 10^{14}$ W cm^{-2} region for three wavelengths of 800, 1200 and 1600 nm, for a pulse duration of 10fs, where the range of Keldysh parameters are 1.06–0.67, 0.71–0.45 and 0.53–0.34, respectively. Thus they lie in the 'accepted' tunnel ionization region. The total ionization probabilities are calculated using the TDSE and from the ADK model. As the laser intensity is increased, the error from the ADK model increases. The error of the ADK model at higher intensities have been addressed earlier. For an atomic hydrogen target in a static field, the ionization rate can be calculated 'exactly' [20–22], since the Schrödinger equation is separable in parabolic coordinates. Using accurate static ionization rates, we calculated the ionization probability for each of the three lasers. The results are shown by the black dash-dotted lines in figure 3. We confirmed numerically that the ionization probability for the 10 fs pulses at fixed peak intensity is independent of the wavelength. For such short pulses, the ionization probability are obtained by integrating ionization rates over the static electric field for each laser pulse. It is interesting to note that the TDSE results for all three wavelengths agree better with the probability calculated using the static ionization rate than with the rates obtained from the ADK model, and the agreement improves with the increase of wavelength. This result clearly supports the adiabatic approximation used in describing laser–atom
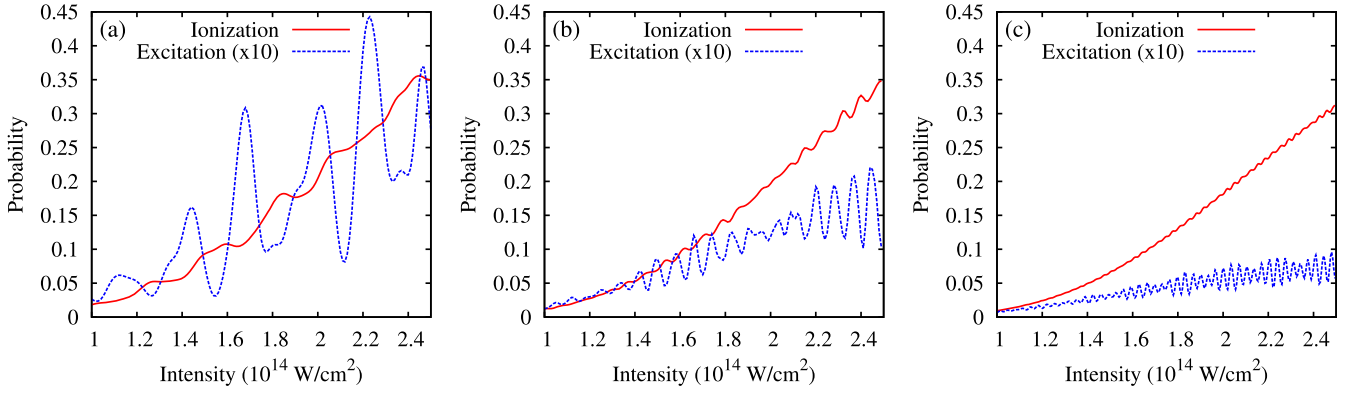
Figure 4. Comparing excitation (multiplied by 10) and ionization probabilities vs intensity for 800 (a), 1200 (b) and 1600 (c) nm wavelengths. The pulse duration is 10 fs.

interaction using mid-infrared lasers. The discrepancy between the ADK model and the TDSE at higher intensities originates from the error in the ADK model in approximating the static field ionization rates. This error can be corrected using an asymptotic series [23].

We stress that static field ionization rates for a one-electron atomic or molecular system can be calculated accurately using the so-called complex rotation method [24, 25]. These data can then be fitted using the method of Tong and Lin [18] to correct the errors of the ADK theory at higher intensities. However, calculations of static field ionization rates for multielectron atoms and molecules are much more difficult, as are TDSE calculations. In such situations, an absolute ionization probability or rate would be difficult to obtain from theoretical calculations. However, differential data such as HHG and photoelectron spectra can still be investigated, even though the precise initial ionization rate is not known accurately.

3. Oscillation of total ionization and excitation probabilities vs laser intensity

In figure 4, we show the total ionization and total excitation probabilities vs laser intensities for the three wavelengths. The excitation probabilities have been scaled up by a factor of ten. Within the same intensity region of $1.0 - 2.5 \times 10^{14}$ W cm^{-2}, the total ionization probabilities do not differ much for the three wavelengths, but the total excitation probability drops by a factor of eight when the wavelength is increased from 800 nm to 1600 nm. The TDSE results for the total ionization probability show small modulations with intensity, more visible for 800 nm than for 1600 nm pulses. The modulations are much stronger for the total excitation probabilities, and the rate of oscillations for longer wavelength is much faster. For 800 nm lasers, these oscillations have been identified to be associated with channel closings in Li *et al* [16], as well as in earlier works by Popov *et al* [26, 27] and others [28, 29] using an approximate model hydrogen atom.

To illustrate these oscillations more clearly, as shown in Li *et al* [16], we first obtained a smoothed probability curve P_{sm} (I) vs the intensity. For excitation, we plot the ratio $P(I)/P_{sm}(I)$. For ionization, we plot $5 \times [P(I)/P_{sm}(I) - 1] + 1$. The out-of-phase oscillation between the two probabilities vs intensity becomes very easy to discern for 800 nm laser, as shown in figure 5(a). In Li *et al* [16], as well as in other earlier references [26–30], these oscillations were found to be associated with channel closing as the laser intensity is increased. According to multi-photon ionization theory, the photoelectron energy is given by $E = n\hbar\omega - (I_p + U_p)$ if the atom absorbs n photons, where I_p is the ionization potential and U_p is the ponderomotive energy. Thus for the lowest ATI peak, its position is shifted closer to the ionization threshold as the intensity is increased. As the intensity increases beyond the n-photon channel closing threshold, one can anticipate that the first 'ATI' peak shifts to below-the-threshold energies where excited states are located. To see this continuous evolution, it is best to express the excitation probability P_n of a given state n in terms of the excitation probability density defined by $dP_n/dE = P_n(dn/dE) = P_n n^3$, since $E(n) = -1/(2n^2)$ for atomic hydrogen [31, 32]. An example of such a plot is shown in figure 6. It shows how the first ATI peak shifts gradually into the energy region where excited states are located. Thus, the decrease in the ionization probability is associated with the increase of excitation probability. Between two channel-closing thresholds, each probability will go through one maximum and one minimum, and they are out of phase. As the wavelength is increased, the oscillation becomes faster—proportional to the cubic power of the wavelength. We comment that it takes 13–19, 27–46 and 49–95 photons, respectively, to ionize H with laser intensities of $1.0 - 2.5 \times 10^{14}$ W cm^{-2} for the three wavelengths considered. Thus, even in the deep tunneling ionization regime, features of MPI are still present in the excitation and ionization spectra.

4. Distribution of Rydberg states and stabilization of atoms in intense laser fields

We next consider the distribution of excited states vs laser intensity. Since the ATI peaks shift to lower energy as the
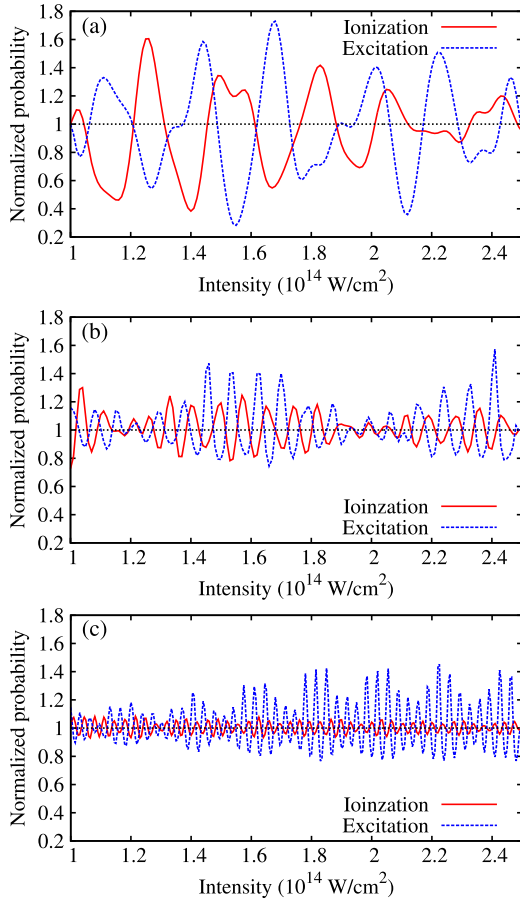
Figure 5. Comparing total excitation and ionization probabilities vs laser intensity showing out-of-phase modulations. The scaling is described in the text and the wavelengths of (a), (b) and (c) are 800, 1200 and 1600 nm, respectively.
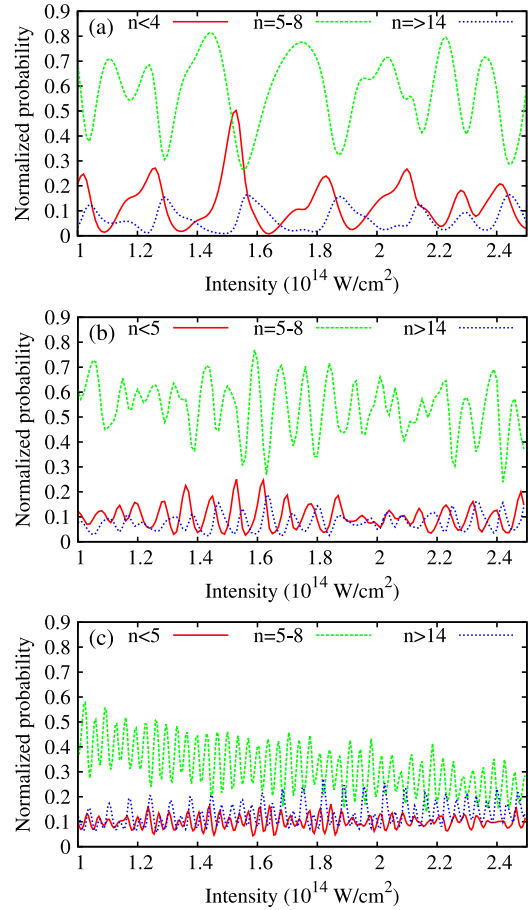

Figure 7. Normalized excitation probabilities for three ranges of principal quantum numbers vs laser intensities for wavelengths of 800 (a), 1200 (b) and 1600 (c) nm, respectively.
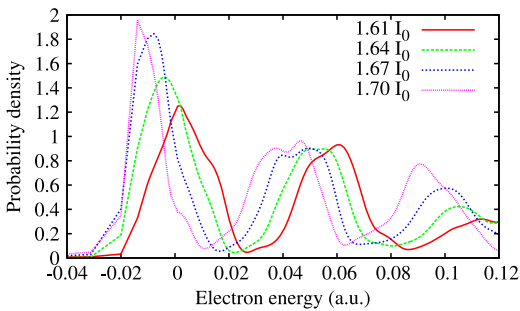

Figure 6. Comparing excitation probability density and ionization probability density to show that the first ATI peak shifts to below-the-threshold region as the laser intensity is increased beyond the channel closing threshold. Data are for 10 fs, 800 nm laser pulses with intensities of 1.61, 1.64, 1.67 and $1.70I_0$, where $I_0 = 10^{14}$ W cm^{-2}. Reprinted with permission from Li *et al* [16]. Copyright (2014) by the American Physical Society.

laser intensity increases between two channel closing thresholds, one expects that the distribution of excited states will move towards lower n values as the intensity is increased. Thus one also expects oscillation in the distributions between higher n and lower n states. In figure 7(a), the populations of $n < 4$, $n = 5 - 8$ and $n > 14$ states are displayed vs laser intensity. In the figure, the total excitation probability is

normalized to 1.0 at each intensity. We note that the $n = 5 - 8$ total probability tends to be dominant. Its intensity dependence shows modulations in accordance with the occurrence of successive channel closings. We also note that the $n < 4$ distributions tend to be out of phase with the $n = 5 - 8$ distributions. Similar distributions for the 1200 and 1600 nm lasers can also be seen in figures 7(b) and (c).

From figure 4, we note that the total excitation probability for an 800 nm laser is of the order of a few percent. The excitation fraction decreases with the increase in wavelength of the driving laser. It is interesting to comment on how a loosely bound electron in the Rydberg states 'survives' the strong electric field of an intense laser.

One common (but wrong) way of questioning the stability of high-n states (Rydberg states) is based on the tunnel ionization picture. The argument is that the electron in these highly excited states lies way above the potential barrier formed by the combined Coulomb field and the 'instantaneous' static electric field of the laser, so that the electron can easily escape from the atom. However, this argument is incorrect since the 'orbiting' period of a Rydberg electron is much longer than the optical period of the laser, thus a static tunnel ionization model is not applicable to Rydberg electrons. In fact, since a Rydberg electron does not move much
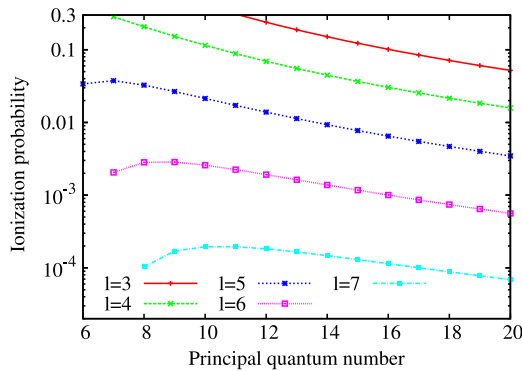
Figure 8. The ionization probability for the absorption of one 800 nm photon for a flat-top pulse of 5 fs duration at the intensity of 10^{14} W cm^{-2} calculated using first-order perturbation theory. Note that the probability scales linearly with laser intensity and pulse duration in this model.

over one optical period, it sees an averaged zero electric field from the laser. Thus, the Rydberg states are stable against intense infrared laser fields.

A semi-quantitative argument to support the stability of Rydberg states against intense laser fields is to view the problem from the multi-photon ionization picture. A Rydberg electron in a state with n greater than 4 can be ionized by absorbing one near- or mid-infrared photon. When this happens, the electron will appear as a Freeman resonance in the photoelectron spectra. If the ionization rate is faster than the formation rate, then the Rydberg state would not survive the laser pulse. However, the ionization rate of a Rydberg electron depends on the quantum numbers, n and l. In the last section, we have shown that the dominant ns in the excited states tend to be in the range 5–8. In Li *et al* [16], it was shown that these excited states have angular momentum quantum numbers greater than 4 or 5. Unlike lower-n and lower-l states that can be efficiently tunnel-ionized, these high-n, high-l Rydberg states (which can be estimated from the corresponding Keldysh parameters) can be destroyed only through multiphoton ionization, including single-photon ionization if it is energetically possible. However, in such states the electron is located far away from the nucleus and behaves almost like a free electron, which does not absorb a photon. As an estimate, we show in figure 8 the ionization probability over 5 fs for a peak intensity of 10^{12} W cm^{-2}. The rates were calculated using first-order perturbation theory for the absorption of a single photon by a monochromatic 800 nm laser. When the probability in the figure is less than, say, a few percent, we can expect that this particular Rydberg state would not be efficiently destroyed by the laser. Thus high-n, high-l Rydberg states will survive the laser field once they are formed. In fact, excited states have been observed in earlier experiments with longer pulses [33, 34].

The discussion above demonstrates that neutral atoms can exist in intense laser fields if the electrons are in high Rydberg states. While infrared lasers can absorb many photons from tightly bound electrons near the nucleus, once the bound electron is promoted to higher excited states, the slow excited electron cannot follow the fast oscillating laser field

and thus they are stable. This is the simple mechanism for explaining the observation of high Rydberg states or the observation of neutral atoms in an intense laser field. Our interpretation here differs from other models like frustrated tunnel ionization [10], interference stabilization [11–13] or the ionization suppression model [14, 15], where more complicated mechanisms have been proposed to explain the same phenomena.

5. Summary

We studied the excitation of Rydberg states and ionization in atomic hydrogen in the presence of an intense short laser field. By analyzing the results obtained from solving the time-dependent Schrödinger equation, we show that the total ionization probability vs laser intensity shows small modulation. This modulation has been traced to be connected to channel closing in the multi-photon ionization picture. The total excitation probability is found also to show strong modulation vs laser intensity. The two oscillations are out of phase, which can be understood by comparing the excitation probability density with the ionization probability density. At the onset of a new channel closing, the first ATI peak 'dives' below the ionization threshold, thus populating high Rydberg states at the expense of reducing photoelectron yields. These high Rydberg states have large orbital angular momentum quantum numbers and thus they are not further ionized by absorbing another photon. They would appear as high Rydberg states at the end of the laser pulse. While the calculations were carried out in atomic hydrogen, the phenomena presented here are general, for any atomic or molecular targets. This work explains why atoms or molecules are not fully ionized, even at extreme high laser intensities. We also discussed the relevance of other ionization suppression models. Our results demonstrate the 'co-existence' of multiphoton ionization features in the strong tunnel ionization regime, even for short, few-cycle pulses.

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