Dynamic Stabilization of Hydrogen in an Intense, High-Frequency, Pulsed Laser Field

Kenneth C. Kulander, Kenneth J. Schafer, and Jeffrey L. Krause
Physics Department, Lawrence Livermore National Laboratory, Livermore, California 94550
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We present calculations of ionization rates, trapping probabilities, and electron densities for a three-dimensional hydrogen atom in an intense, high-frequency, pulsed laser field. Under certain conditions, we find that hydrogen has a significant probability of surviving the ramp of the laser field to intensities at which ionization is strongly suppressed. We examine the dynamics of this process and discuss a method to observe it experimentally.

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In the limit of asymptotically high laser frequency, an atom stabilizes with respect to ionization, and its electronic wave function localizes near the nucleus in a well-defined volume.[1] Recent calculations have predicted that the ionization rate of a hydrogen atom in its ground state increases to a maximum and then decreases with increasing laser intensity, provided the frequency is high enough.[2] These predictions are based on solutions of the nonrelativistic Schrödinger equation in a classical, linearly polarized laser field with a constant intensity. Several major questions concerning the dynamics and detection of localization could not be addressed by these previous calculations. For example, in a realistic laser pulse with a finite frequency, what is the probability that an atom survives the ramp of the laser field through intensities at which it ionizes rapidly? What is the form of the stabilized state? If such a state is created in an experiment, how can it be observed?

One-dimensional calculations of two-photon (field-free) ionization of a model atom have provided partial answers to these questions.[3] In this paper, we present the first three-dimensional results of quantum-mechanical calculations of ionization rates, trapping probabilities, and electron densities for a hydrogen atom in an intense pulsed laser field. We use a pulse consisting of a $\sin^2$ ramp of the field over 1–10 optical cycles, followed by 10–100 cycles of constant intensity. Under certain conditions, we find that a very significant fraction of the wave function survives the rapid turn-on of the laser field and remains trapped in a state localized near the nucleus. During the constant-intensity part of the pulse, we observe a reasonably well-defined exponential decay of the charge density remaining in the vicinity of the nucleus. As the intensity increases, the survival probability *increases,* and the ionization rate of the localized state *decreases* dramatically. We examine the dynamics and composition of the localized state, and calculate its photoemission spectrum as a possible experimental signature of stabilization.

The calculations reported here were performed with the finite-difference codes described elsewhere.[4] The codes were modified to use the velocity gauge, which we found to be more efficient computationally than either the length gauge or the acceleration gauge for conditions of high intensity and frequency. The calculations proceed by defining a grid in space with boundaries well removed from the nucleus. The dimensions are several times larger than $\alpha_0 = E/\omega^2$, the amplitude of oscillation of a free electron in a laser field of strength E and frequency ω. To prevent reflection of the wave function from the edge of the grid, we apply a smooth cosine mask function confined to a moderately narrow band near the boundaries after each integration step to force the electron density to zero at the edges of the grid.[4,5] Since it is unlikely that electrons very far from the nucleus will return to the atom, we assume that electron density removed by the mask function corresponds to ionization. Removing the ionized portion of the wave function causes the overall norm of the wave function remaining on the grid to decrease with time. If the norm decreases exponentially during the constant-intensity portion of the laser pulse, an ionization rate can be defined as the slope of the logarithm of the norm versus time. In earlier studies at lower intensities and frequencies,[4] we found that an ionization rate defined by the decay of the field-free ground state agreed with the rate defined by the decay of the norm. Under the conditions studied here, the stabilized state contains only a very small fraction of the field-free ground state and no sensible rate can be obtained from this projection. As we shall illustrate below, the trapped state bears no resemblance to the field-free ground state. Indeed, after the laser pulse is over, the remaining electron density is broadly distributed over the entire bound-state manifold.

We would like to emphasize that the calculations presented here are nonrelativistic. Some evidence exists from classical calculations that relativistic effects may inhibit stabilization.[6] More work is needed to fully understand the consequences of relativity in very intense laser fields. We note that the nonrelativistic classical results predict that stabilization in 3D does not occur until frequencies much higher than those predicted by our quantum-mechanical calculations.

Figure 1 shows the time-dependent norm of the wave

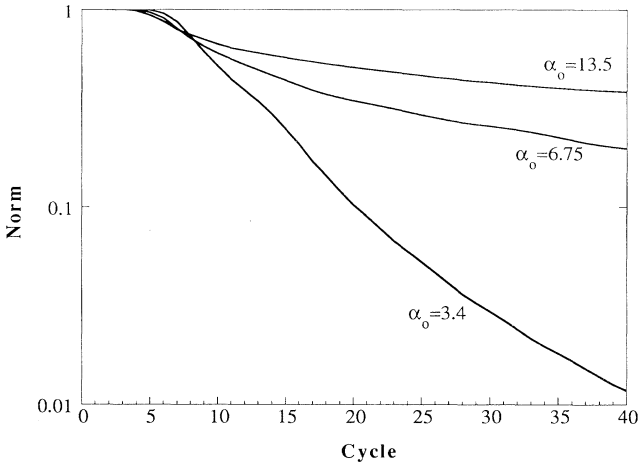

FIG. 1. Time-dependent survival probability for a hydrogen atom in a laser field of frequency 1 a.u., for $\alpha_0=3.4$, 6.75, and 13.5 a.u. The intensity was ramped to its maximum value over 5.25 cycles with a $\sin^2$ pulse.

function for a hydrogen atom in a laser field of frequency 1 a.u. (27.21 eV, twice the field-free binding energy) and intensities of 4×10^{17}, 16×10^{17}, and 64×10^{17} W/cm^2, corresponding to $\alpha_0=3.4$, 6.75 and 13.5 a.u., respectively. These calculations used a pulse consisting of a 5.25-cycle $\sin^2$ ramp followed by 95 cycles of constant intensity. In all three cases, the norm decreases approximately linearly on this semilogarithmic plot during the later cycles of the pulse. The ionization rates derived from the last 50 cycles of the pulses are 8.7×10^{14}, 1.5×10^{14}, and 0.37×10^{14}/s, respectively. At the end of 40 cycles, the norms are 0.01, 0.20, and 0.38. Clearly, the effects of stabilization are dramatic. The highest-intensity case results in an ionization rate slow enough to allow a considerable population of stabilized atoms to exist for physically realizable pulse lengths (i.e., tens to hundreds of optical cycles).

The ionization rates calculated in this work are in general agreement with those calculated via a nonperturbative Floquet approach for the hydrogen atom in its ground state.[2] For example, in a laser field of frequency 1 a.u., we obtain a peak rate of 1.3×10^{15}/s at $\alpha_0=1.2$, compared to a peak rate of 5.4×10^{15}/s at $\alpha_0=1.2$ as calculated with the Floquet method.[2(b)] Once the intensity has increased to the point that $\alpha_0\gg1$ and the stabilized state has formed, the ionization rate varies slowly with α_0. Further increases in intensity cause only small decreases in the rate.

The results in Fig. 1 are sensitive to the slope and duration of the ramp of the laser pulse. If the ramp is too abrupt, negligible trapping occurs. For instance, at $\alpha_0=13.5$, a ramp of 1.25 optical cycles results in a norm of less than 0.01 after 40 cycles. If the laser is turned on too slowly, rapid ionization occurs during the ramp and very little trapped population remains by the time the pulse has reached its maximum intensity. We have found, however, that trapping is efficient for a range of laser pulses. For example, a ramp of 10.25 cycles results in a norm of 0.25 after 40 cycles, compared to 0.38 for a 5.25-cycle ramp.

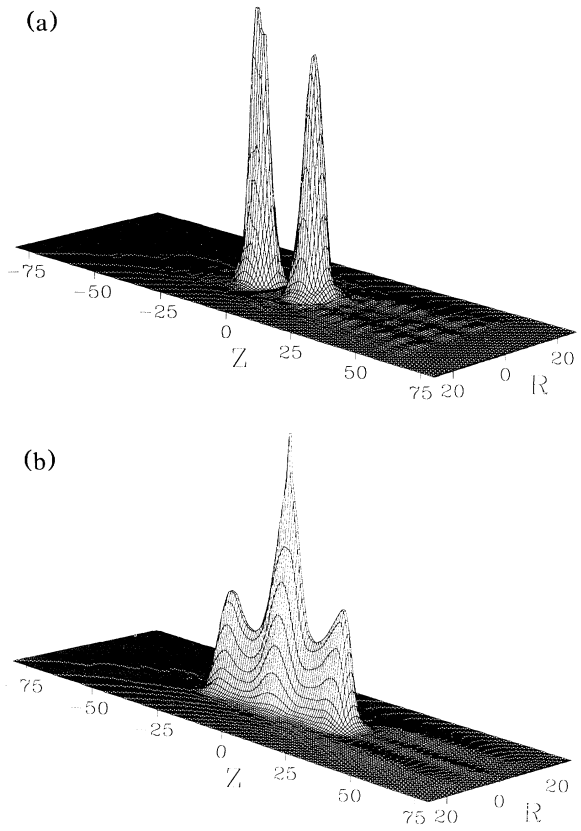

FIG. 2. (a) Snapshot of the electron density at the beginning of the 21st optical cycle for a hydrogen atom in a laser field of frequency 1 a.u. and $\alpha_0=13.5$. The intensity was ramped to its maximum value over 5.25 cycles with a $\sin^2$ pulse. (b) Time-averaged electron density for the conditions in (a), averaged over the 21st optical cycle.

In the high-frequency limit, the localized state is an eigenstate of the time-averaged potential in the Kramers-Henneberger (KH) frame.[7] As α_0 increases, the ground state of this potential splits into two peaks at $z=\pm\alpha_0$, where z is the direction of polarization.[1] At large α_0, the peaks are well separated, and the "dichotomy" is complete. In the laboratory frame, the trapped state exhibits the same shape as in the KH frame, but oscillates with an amplitude of α_0 in phase with the laser field.[1(a)]

To test the robustness of the theory based on asymptotically high frequency and constant laser intensity, we have examined the time evolution of the trapped state created at a *finite* frequency in a *pulsed* laser field. Figure 2(a) shows a snapshot of the time-dependent electron

density at the beginning of the 21st optical cycle for a hydrogen atom in a laser field with a frequency of 1 a.u., $\alpha_0=13.5$, and a 5.25-cycle $\sin^2$ ramp. The density shows two peaks, separated by approximately $2\alpha_0$, with slightly different heights. Over the course of a *single* optical cycle, the general shape of the density is preserved. The entire structure oscillates in phase with the driving field with an amplitude of α_0. When the probability density is time averaged over a single optical cycle, as shown in Fig. 2(b), it has three peaks, in agreement with the predictions of Pont and co-workers.[1]

Following the evolution of the density over *many* optical cycles, we find that its shape changes gradually but dramatically. After fifteen additional cycles, the density evolves to a form with a large central peak and two smaller, asymmetric side peaks. The overall width of the function remains $2\alpha_0$. After fifteen more cycles, the shape shown in Fig. 2(a) reappears. The period of this recurrence is in quite good agreement with that predicted by the energy difference of the two lowest σ_g states ($1s$ and $2s$) of the time-averaged potential.[1(b)] A similar recurrence is found at $\alpha_0=15$ a.u., with a period of 42 cycles, again in good agreement with the $1s$-$2s$ splitting. These recurrences suggest that the localized state created during the ramp of the laser field is a *linear combination* of dressed states of the time-averaged potential, consisting predominantly of the two lowest σ_g states. A small admixture of the lowest σ_u ($2p$) state yields the slight asymmetry in the peak heights of Fig. 2(a). We discuss below the significance of a component of odd symmetry in the trapped wave function. The excited dressed states ionize more slowly than the ground state.[2(b)] This may account for the differences in the ionization rates for the hydrogen atom calculated by the Floquet method and in this work.

The composition of the localized state depends critically on the ramp of the laser field. If the ramp is too abrupt, its large bandwidth causes a nonadiabatic population of a large number of states, most of which lie in the continuum. It may be that the best way to create a state at high α_0 which contains only a few low-lying eigenstates of the time-averaged potential (and hence a state with a clean dichotomy) is to use a moderately fast ramp to about $\alpha_0=10$, followed by a slower ramp to higher α_0.

Finally, we address the problem of observing the stabilized state. We believe that the spatial distribution of intensities present in a laser focus will make it very difficult to construct an experiment in a volume of gas which can measure a decrease in the ionization rate with increasing laser intensity. However, photoemission from the stabilized state, which might include high harmonics, offers an experimentally accessible means to detect localization.

To investigate this possibility, we calculate the induced, time-dependent dipole of the localized wave function. The square of the Fourier transform of the dipole, shown in Fig. 3, is proportional to the atomic photoemission spectrum.[8] We performed the Fourier transform over cycles 21–51, or one period of the recurrence caused by the $1s$-$2s$ mixing. Strong harmonics at least to fifth order are visible above a broad background. One surprising aspect of Fig. 3 is the presence of *even* harmonics. An isolated atom in a continuous-wave laser field has inversion symmetry, so even harmonics are forbidden by parity conservation. However, as we mentioned previously, the localized wave function contains components of odd symmetry. This occurs because the ionization rate reaches a maximum within an order of magnitude of the laser frequency and then decreases during the ramp of the pulse. Therefore, the rate during the first half of an optical cycle can be very different than the rate in the second half of the cycle. During each half cycle, the ionized flux leaves the vicinity of the atom along the direction defined by the instantaneous electric field. This can be seen in Fig. 2(a), where a small amount of ionizing flux is evident in the positive z direction. Asymmetric ionization leads to a nonsymmetric medium, which is capable of sustaining even harmonics. We find that a slower ramp, which causes more symmetric ionization, appreciably diminishes the relative strengths of the even harmonics. Figure 3 is, to our knowledge, the first evidence of even harmonics in a single-atom spectrum. Note that we do not propose the observation of even harmonics alone as a signature of stabilization. The inversion symmetry of the medium may also be broken by macroscopic inhomogeneities induced by the short pulse laser. These two sources of even harmonics can be distinguished if the density dependence of the harmonic conversion could be measured.[9]

The harmonic radiation from atoms in the focal volume phase matches to produce a strong coherent signal in the forward direction, which should be an observable signature of stabilization. An ideal experiment would measure the harmonic production as a function of

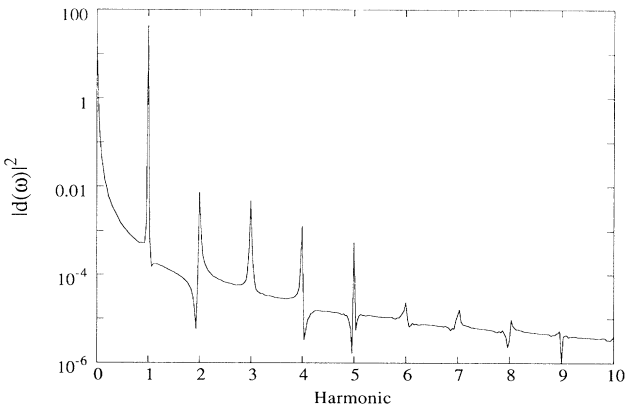

FIG. 3. Photoemission spectrum for the conditions of Fig. 2.

peak laser intensity. We predict that the harmonics, if they are visible at low peak intensity, will disappear as the intensity increases to the point that the medium is fully ionized and then reappear when the peak intensity reaches the stabilization regime.

This paper has concentrated on dynamic stabilization of the hydrogen atom. The conditions required to stabilize the ground state of hydrogen are difficult to realize experimentally at present. However, these calculations have enabled us to identify the aspects of the problem that are critical in designing an experiment to verify the theoretical predictions. First, the laser pulse must pass through the region near $\alpha_0 = 1$ as quickly as possible. This does not necessarily imply the use of ultrashort pulses, but does require pulses which ramp through a relatively narrow intensity range in at most a few optical cycles. Second, stabilization becomes less effective as the number of photons required for ionization increases. A system with a binding energy lower than H would allow the use of longer-wavelength photons, and hence dramatically lower peak intensities.

We would also like to point out that stabilization may exhibit dimensionality effects. Pont and co-workers[1] predict a trapped wave function with a width of $2\alpha_0$, which was verified in this work (Fig. 2). Recent quantum 1D calculations show a function with a spacing between the peaks of α_0.[3] A classical analysis of the 1D case also predicts spacings of α_0.[10] These results are not inconsistent with the superposition state observed here. The peak spacings depend on which states are excited by the ramp. Part of the difference between the 1D and 3D calculations may be that the 1D calculations were performed for the case of two-photon ionization. We have found that one-photon ionization leads to a cleaner dichotomy (that is, a superposition containing mostly low-lying states) than the two-photon case. We have observed that the structure of the trapped state is even more sensitive to the rate and shape of the ramp in 1D than it is in 3D.

We conclude by surmising that stabilization occurs because the localized state has very little amplitude in regions in which photoabsorption is allowed. Momentum conservation forces photons to be absorbed only near the nucleus. As the frequency increases, the volume around the nucleus in which transitions can occur becomes very small. Increasing the intensity causes the spatial extent of the trapped wave function to increase, which lowers the density near the nucleus, and decreases the ionization rate. In 3D, it is easier for an electron to avoid regions near the nucleus than it is in 1D, so it is not surprising that the 3D atom shows a comparable or greater tendency to localize than the 1D atom.[3,10]

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