Dynamics of Atomic Ionization Suppression and Electron Localization in an Intense High-Frequency Radiation Field

Q. Su and J. H. Eberly
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627

J. Javanainen
Department of Physics, University of Connecticut, Storrs, Connecticut 06268
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We present the results of high-frequency intense-laser numerical multiphoton experiments on a model atom. These results indicate both suppression of ionization and localization of the atomic electron even after a smooth laser turn on.

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The Kramers-Henneberger (K-H) transformation[1] has been used by Gersten and Mittleman[2] and by Gavrila and co-workers[3] to infer stabilization of the hydrogen atom under intense-laser excitation. The K-H transformation allows one to view the H atom from the moving coordinate frame of a completely free electron responding to a monochromatic laser field. The K-H frame is therefore expected to be useful if the laser field is comparable to the Coulomb field in determining the electron response. In the K-H frame the Coulomb potential becomes what we will call the K-H potential:

$$\frac{-1}{|\mathbf{r}|} \rightarrow \frac{-1}{|\mathbf{r}+\alpha(t)|} = \frac{-1}{|\mathbf{r}+\alpha_0 \sin\omega t|}, \tag{1}$$

where the laser field has been taken to be $\mathbf{E}=\mathcal{E}\sin\omega t$ and α_0 is the amplitude of free-electron response, and is given by

$$\alpha_0 = -e\mathcal{E}/m\omega^2. \tag{2}$$

If the frequency of oscillation of the laser field is assumed asymptotically high, then it is valid to discard all but the time average of the K-H potential (1). The Schrödinger Hamiltonian remains real under the K-H transformation, and is obviously time independent when time averaged, so one is led to the prediction[2,3] that all energy eigenstates will be stable. In other words, if the frequency is high enough to justify the time average, the atom will make no transitions, no matter how strong the field, and transitions leading to photoionization in particular will be eliminated. This is the meaning of stabilization.

Pont *et al.*[3] have obtained solutions numerically for a variety of energy eigenstates in the time-averaged K-H potential. This is equivalent to making a Floquet expansion of the wave function and keeping only the zero-photon term. One of the striking features of their solutions is a dramatic splitting of the electron probability distribution into a clear two-peaked or "dichotomous" distribution as soon as the laser intensity is large enough to make $|\alpha_0|$ greater than about 10 bohr radii. This can be expected classically because the most probable position for the electron in the K-H potential is at the two extremes of its excursion, at $\mathbf{r}=\pm\alpha_0$ where it momentarily has zero velocity. Thus Gavrila and co-workers[3] predict localization as well as stabilization.

Predictions of atomic stability (zero-level widths) under high-frequency (laser) excitation are relatively new in atomic physics,[4] although a strong analog exists in resonance physics in the concept of motional line narrowing. High intensity of the laser field motivates one to search for a viewpoint (the K-H picture) that emphasizes the effect of the laser field while still retaining some of the binding of the Coulomb field. In this respect the K-H picture may be an improvement on the better-known Keldysh picture, in which Coulomb effects are included only through the use of the zero-intensity Coulomb wave function to describe the initial state.[5]

Despite the attractiveness of the K-H picture, and the detailed calculations of the properties of the stabilized K-H atom that are appearing,[3] little progress has been made in understanding the *dynamics* of high-frequency ionization. Therefore, some interesting questions remain unanswered, both quantitatively and qualitatively. (1) Can a stabilized state be reached following a smooth turn on of the laser pulse? (2) What degree of stability can be expected—can it be as great as 10%, for example? (3) Given a specific laser frequency, is there a threshold intensity at which the K-H picture becomes valid in a practical sense? (4) What value of laser frequency is sufficiently high to guarantee a finite degree of stability? And (5) how rapid is the residual ionization due to non-zero-order Floquet terms that are ignored by the time-averaged K-H potential?

It is the purpose of the present Letter to address these open questions. We will do so by presenting the results of a series of numerical experiments. We have carried out these experiments on a one-dimensional model hydrogen atom which we have subjected to high-frequency intense-laser pulses turned on over 5.25 and 15.25 optical cycles using the smooth turn-on function $\sin^2(\pi t/T)$. We will present results for a wide range of laser peak field strengths, $\mathcal{E}=0.01$–5.0 times the atomic unit of field strength, corresponding to laser intensities between 3.5×10^{12} and 9×10^{17} W/cm^2. The ionization threshold of the model atom is $E_{\text{ion}}=18.21$ eV and the energy of the laser photons is $\omega=14.13$ eV.

The characteristics of the model atom we use have been reported before,[6] as well as the numerical methods[7] employed to obtain time-dependent solutions $\Psi(x,t)$ to Schrödinger's equation. The bare atom is characterized by the long-range, asymptotically Coulombic binding potential $V(x) = -1/(1+x^2)^{1/2}$, and the numerical eigenvalues and eigenfunctions we have calculated for it share many of the properties of real atoms: a Rydberg series near threshold, parity as a good quantum number, satisfaction (to five significant figures) of the dipole sum rule, etc. In addition, agreement between previous time-dependent high-intensity phenomenological predictions[8] of the model and either laboratory experiments or other model calculations has been uniformly good.

Given this model atom, and the laser parameters mentioned, our results are most directly applicable to two-photon ionization (before pondermotive shifts) of an atom with an energy spectrum similar to that of hydrogen, but an ionization potential about 50% greater. We will address only qualitative questions (e.g., does stabilization exist? can stability be reached via a smooth laser turn on?, etc.), so the details of the differences between the model and any specific real atom or real laser are not expected to be important in first approximation.

Our first results are shown in Fig. 1. We show the ionization probability

$$P(t) = \int dW \,|\langle W|\Psi(t)\rangle|^2 \tag{3}$$

(integrated over more than 1500 positive-energy eigenstates $|W\rangle$) as a function of time for four values of peak laser field strength $\mathcal{E}$ that are smaller than one atomic unit. Note that the four graphs have very different vertical scales. The figures show that in this intensity range, after adjusting to the 5.25-cycle turn on, the atom exhibits mostly smoothly growing ionized probability. In Figs. 1(a)-1(c) the post-turn-on linear slopes (ionization rates) are greater in succession by large factors consistent with the expected power law I^2 [these factors are 625 and 16, respectively, between (a) and (b) and between (b) and (c)]. In Fig. 1(d) the field strength is half an atomic unit and while the ionization process remains relatively smooth it is no longer linear. It is then not possible to define a steady ionization rate.

In Fig. 2 we show our results for stronger fields. Figure 2(a) repeats Fig. 1(d) for cross reference. All four graphs have the same vertical scale. The absence of linear ionization growth is seen in all four cases and begins at about $|\alpha_0| \approx 1$ and lends support to the K-H theme that α_0 is the key parameter of strong-field ionization. Note also that the behavior in Fig. 2 is suggestive of stabilization. Every curve for $\mathcal{E} = 1.0$ and above shows a tendency to saturate before reaching $P = 1$. Even more interesting, the saturation level appears to be lower for higher field strength. Of course our results do not prove that the atom is truly stable under a long-extended application of the laser field. There may be a small residual ionization rate. However, it is quite clear that the *amount of ionization is anomalously low, and becomes lower at higher field strength.* This is what is sensibly meant by stabilization (the suppression of ionization) in a strong-field environment.

Our model approach to the suppression question has deficiencies, of course, mostly connected with the one-dimensional character of our model, but these deficiencies are quite different from the deficiencies of the K-H Floquet method followed previously.[2,3] In particular, our calculations are for a concrete and finite ω rather than for an asymptotically high frequency, and they are for a laser field with a finite quasiadiabatic turn on rath-

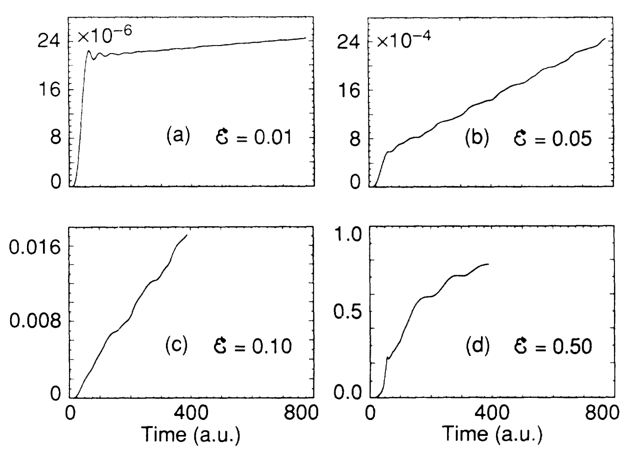

FIG. 1. Electron ionization probability as a function of time for four moderately high laser intensities [$I = (3.5\times10^{12})$-(9×10^{15}) W/cm^2]. The laser electric field is turned on gradually over 5.25 cycles, and the peak field strength is noted in each frame. The vertical scales differ greatly from (a) to (d). The full horizontal axis corresponds to about 64 laser cycles at the frequency employed. In (a)-(c) the asymptotic straight-line slopes scale according to the perturbative I^2 law.

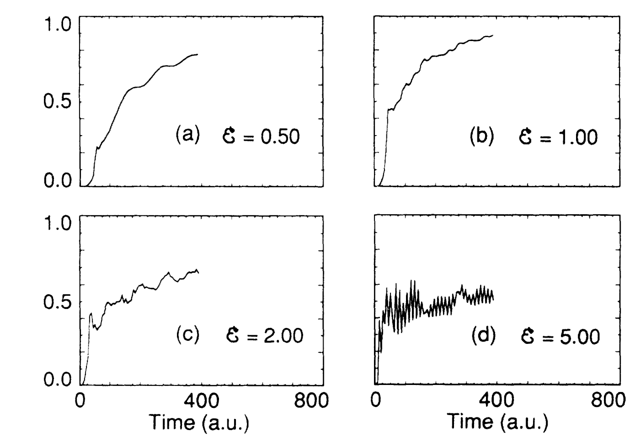

FIG. 2. Electron ionization probability as a function of time for four laser pulses turned on over 5.25 cycles and reaching very high laser intensities [$I = (9\times10^{15})$-(9×10^{17}) W/cm^2]. The vertical scales are the same in each graph. The increasing degree of saturation from (b) to (d) is evident.

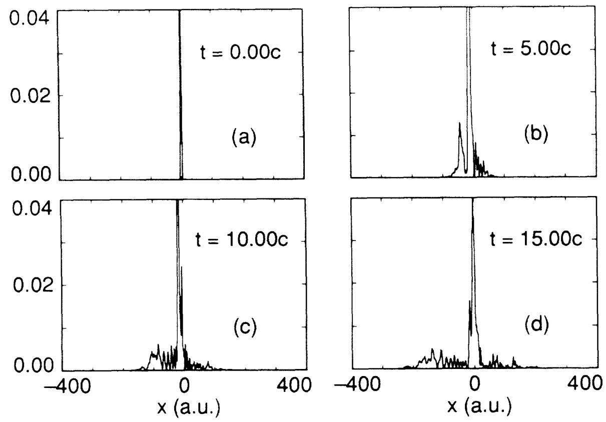

FIG. 3. Electron probability density $|\Psi(x,t)|^2$ vs x near to $x=0$ for four different times (as indicated by numbers of laser cycles since $t=0$). The electron probability distribution near $x=0$ sheds a packet that gradually moves to the left. The laser parameters are those given for Fig. 2(d).

er than for a pure sine-wave field. In effect, our approach, the direct solution of the fully time-dependent Schrödinger equation, includes Floquet terms of all orders. Thus we believe these results give the first indication that stabilization can be expected to persist in the face of various experimental realities and can be realistically pursued in the laboratory.

In view of the various mechanisms that have been proposed to lead to stabilization,[2-4] we can now ask whether our numerical experiments distinguish between them. That is, does the stability ($P<1$) indicated in Fig. 2 conform to a particular type of stability already discussed in the literature? In order to study this question we have also computed electron space-time probabilities from our wave functions. And, as it turns out, these probabilities show (see Figs. 3-5) that we are observing the stability associated with *dichotomous localization* whose static eigenfunctions have been carefully studied by Gavrila and collaborators.[3] The space-time probabilities also provide, incidentally, a check on the results show in Fig. 2 by confirming that the suppressed ionization probability that we computed by the wave-function projection (3) is accompanied by spatial localization of the electron near the nucleus. Suppression of ionization is not just an artifact of gross shifts in the bare eigenlevels $|W\rangle$.

In Fig. 3 we show the electron probability $|\Psi(x,t)|^2$ as a function of x for a variety of early times near the turn on, for the same laser pulse with $\mathscr{E}=5.0$ that gave rise to Fig. 2(d). Figure 3(a) shows that the electron is well localized in its ground state at $t=0$ before the laser pulse arrives, and Fig. 3(b) indicates that a significant packet of electron probability has already begun to move toward negative x at $t=5$ cycles. At $t=10$ and 15 cycles we see that this packet continues to move away. It probably chooses to move to the left because of the minus sign in the classical free-electron amplitude (2).

The effective suppression of ionization seen already in Fig. 2(d) above can be seen by comparison with space-time plots at later times. In Fig. 4 we show $|\Psi(x,t)|^2$ at $t\approx 32$ cycles. The similarity to $|\Psi(x,t)|^2$ at $t=10$ and 15 cycles implies that stabilization has taken effect and ionization has practically ended.

However, the space-time plots show more than confirmation of the suppression of ionization. They allow the unambiguous observation of peak splitting ("dichotomy"). The four individual graphs in Fig. 4, taken a quarter of a cycle apart, also show clear temporal oscillations from Figs. 4(a) to 4(d). Close inspection confirms that these changes, as well as the peak splitting, are quantitatively consistent with the K-H picture of strong-field ionization. In particular, Fig. 4(d) shows the probability at 32.25 cycles on an expanded scale. The peak-to-peak separation is $\Delta x\approx 20$ a.u., closely consistent with our value of $\alpha_0=18.49$ a.u.

We have also computed the results of an experiment identical to the one just described, except that the laser turn on is 15.25 cycles, almost 3 times longer. In this case the major consequence is to alter slightly the character of the localized probability peaks seen in Figs. 4(a)-4(d). The peak amplitude of the localized fraction is still very nearly equal to that shown in Fig. 4. A $P(t)$ curve for the 15.25-cycle turn on (not shown here), corresponding to that shown in Fig. 2(d) for a 5.25-cycle turn on, saturates at about $P\approx 0.7$, indicating that, as expected, longer turn ons will lead to greater ionization and a lower degree of stabilization.[9]

Our results provide answers to a number of outstanding questions in superintense laser-atom physics. They indicate that (1) suppression of ionization, predicted from earlier treatments[2,3] based on the assumption of asymptotically high-frequency laser light, is realizable at finite near-optical frequencies; (2) the threshold laser in-

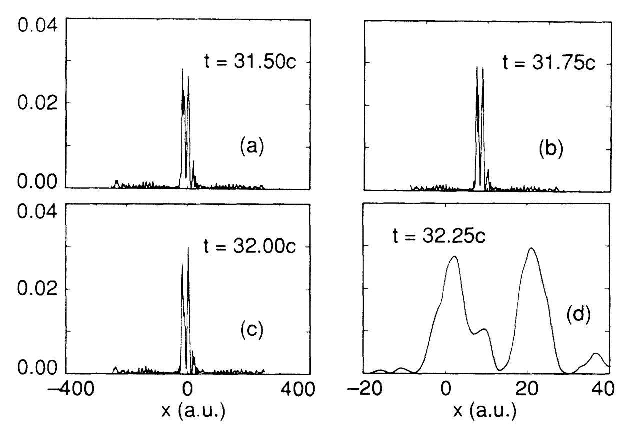

FIG. 4. Same as Fig. 3, except at later times, as indicated. Note "dichotomous" peak splitting and oscillation of peak positions as implied by the K-H viewpoint. (d) is drawn on an expanded horizontal scale to show details near to $x=0$; e.g., the peak splitting is about 20 a.u., consistent with the value $\alpha_0=18.49$ a.u.

tensity for suppression is not unreachably high, a few times above the atomic unit; (3) ionization suppression is accompanied by spatial localization and in some cases by clear dichotomous residual binding; (4) the degree of atomic stabilization can be surprisingly high, even approaching 50%; (5) no residual ionization rate has been measured, but a small residual rate is not inconsistent with our data; and, perhaps most important, (6) these findings all remain valid even in the presence of finite and smooth laser turn ons.

From these findings we can conclude that the K-H picture[1-3] is qualitatively reliable, and that the speculations of earlier workers[1-3] as regards the existence of localized electron dichotomy and the role of the parameter α_0 are semiquantitatively confirmed. Of course, given this confirmation, it is still possible to hold open the question of the most effective explanation for stabilization. It may be that resonance interference within the highly distorted K-H level structure plays a major role. This is consistent with a finding of Kulander[10] and clearly related to the discussions of Ref. 4.

In addition, we have found that the *degree* of stabilization increases with laser intensity (recall Fig. 2). In any event it suggests that a critical intensity exists for a given frequency, a suggestion not possible from the K-H viewpoint, which only implies that any intensity at all will give stabilization if the frequency is high enough.

Our model's parameters are not quite optical, and no high-power source exists for photons that are capable of two-photon ionization of hydrogen. Still, the model illustrates concretely the dynamical response of an atomic electron to a strong finite-frequency radiation field. Past experience[8] indicates a good semiquantitative correlation between our model's predictions and experimental observations. On this basis it appears reasonable to conclude that suppression of ionization and spatial localization under strong-field excitation may be accessible to experimental test with lower-frequency photons and an atom with a correspondingly lower ionization potential. We will present elsewhere an extended account of our results, including associated above-threshold ionization (ATI) spectra.[11]

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