RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 44, NUMBER 7 1 OCTOBER 1991

Observability of atomic stabilization in an intense short pulse of radiation

Marcel Pont and Robin Shakeshaft
Physics Department, University of Southern California, Los Angeles, California 90089-0484
(Received 16 May 1991; revised manuscript received 1 July 1991)

We present results of a numerical integration of the time-dependent Schrödinger equation for hydrogen irradiated by a linearly polarized high-frequency Gaussian pulse, with the atom initially in a state with principal quantum number equal to 3, 4, or 5. We show that stabilization against ionization commences when the ponderomotive and photon energies are roughly equal, and we give a simple explanation of this effect based on momentum-energy considerations. The effect should be observable if the atom is initially in a state with high magnetic quantum number m, and if the energy bandwidth of the pulse is smaller than the binding energy of the atom.

PACS number(s): 32.90.+a, 32.80.Rm

Recent theoretical studies [1] have indicated that an atom exposed to radiation of high frequency becomes stable against ionization for sufficiently high intensities. However, a hitherto unsettled issue is whether this effect is observable in the laboratory with present-day technology. What is the intensity I_{onset} for the onset of stabilization? Does the atom ionize at intensities below I_{onset} if the pulse is reasonably short, say 50 cycles? Does the frequency bandwidth play an important role, and does the spatial inhomogeneity of the intensity mask stabilization? We argue that if the atom is initially in a state with moderately high magnetic quantum number m, stabilization should be observable. We present results of a numerical integration of the time-dependent Schrödinger equation for a hydrogen atom that is initially in the $3s$, $3d$, $4s$, $4p$, $4f$, $5s$, or $5g$ state and that is irradiated by a pulse of linearly polarized high-frequency photons. By "high frequency" we mean that the photon energy $\hbar\omega$ is substantially larger than the unperturbed binding energy $|E_0|$. We show the following. (i) I_{onset} is that intensity for which the ponderomotive energy is roughly equal to the photon energy. We give a simple explanation of this law, based on kinematical considerations. (ii) If the atom is initially in a high m state, say the $5g(m=4)$ state, a large fraction of atoms survives irradiation by a 50-cycle pulse, even for peak intensities above I_{onset}. (iii) For these high-m states, the ionization yield follows a simple scaling law as the pulse duration increases beyond a few cycles. (iv) The pulse must be sufficiently long for stabilization to occur—we speculate that the energy bandwidth must be small compared to the initial binding energy of the atom, for otherwise ionization occurs through Raman transitions to the continuum. (v) Spatial averaging of the ionization yield over the intensity profile does mask the onset of stabilization; however, provided that the confocal parameter is much larger than the atomic sample dimensions, the derivative, with respect to peak intensity, of the measured (spatially averaged) yield is proportional to the unaveraged yield.

Our calculations were carried out by representing the wave function on a complex Sturmian basis [2]. We split the Hamiltonian into two parts, separating the bare atom from its interaction with the radiation, and we used the split-operator method [3] to propagate in time. The method has been described in detail elsewhere [4]. We assume that the pulse has an intensity profile $I(t) = I_0 e^{-(t/t_p)^2}$, with full width at half maximum (FWHM) equal to $2\sqrt{\ln(2)}t_p$. In the following figures we show the probability for ionization at the end of the pulse (i.e., the fraction of atoms that ionize) versus the *peak* intensity I_0 of the pulse. In Fig. 1 we show results for the case where the atom is initially in the $3s$, $4s$, or $5s$ state, ω=0.2 a.u. (about 5.4 eV, a factor of 3.6 larger than the binding energy of the $3s$ level), and the FWHM of the pulse is five cycles. The ionization probabilities rise linearly with increasing I_0 in the perturbative regime, but begin to level off as I_0 approaches 1×10^{15} W/cm^2, when the ponderomotive energy at the peak intensity is equal to $\hbar\omega$. As I_0 increases further, the 3s and 4s ionization probabilities decrease slowly, and the 5s ionization probability only barely increases. Thus the atom becomes stable against ionization once I_0 exceeds $I_{\text{onset}} = (\hbar\mu c/2\pi e^2)\omega^3$, with e and μ the electron charge and mass.

This criterion for the onset of stabilization, which has been argued from different points of view [5], has been illustrated previously by results of Floquet calculations [2], and is supported not only by the results shown here but also by many other results (at various frequencies) that we have obtained within the time-dependent framework. We now sketch a derivation which clearly shows that the origin of stabilization is kinematical. Note first that during each atomic orbital revolution, the nucleus (or, more, generally, the atomic core) can easily communicate to the electron a momentum comparable to the characteristic atomic orbital momentum $\sqrt{2\mu|E_0|}$. However, if the electron absorbs N high-frequency ($\hbar\omega \gg |E_0|$) photons, it emerges with a drift momentum of magnitude $p_{\text{dr}} \approx \sqrt{2\mu N\hbar\omega} \gg \sqrt{2\mu|E_0|}$. Since photons carry no momentum (in the dipole approximation), and since the atomic orbital period is much longer than the cycle time $T_{\text{cyc}} \equiv 2\pi/\omega$, the electron does not have time,

during one cycle, to acquire such a large drift momentum through a series of "soft" collisions (in contradistinction [6] to the low-frequency regime, where $\hbar\omega \ll |E_0|$). Rather, high-frequency ionization takes place through a "hard" collision [6], which we picture as follows. The electron, driven by the electric field $\mathrm{Re}Fe^{-i\omega t}\boldsymbol{\epsilon}$, is incident on the nucleus with the quiver momentum $\mathbf{p}_{\mathrm{quiv}}(t) \equiv -\mathrm{Im}(eF/\omega)e^{-i\omega t}\boldsymbol{\epsilon}$, at time $t = t_{\mathrm{col}}$, say. Assuming that the collision duration Δt_{col} is small compared to T_{cyc}, the collision is sudden on the time scale of the field oscillations and is therefore elastic (c.f. scattering in the presence of a low-frequency field [7]). *Immediately after* the collision, the electron has instantaneous momentum $\mathbf{p}_{\mathrm{quiv}}(t_{\mathrm{col}}) + \mathbf{p}_{\mathrm{dr}}$ (the momentum transfer is $\mathbf{p}_{\mathrm{dr}}$) and energy conservation implies that $\Delta E(t_{\mathrm{col}}) \equiv [|\mathbf{p}_{\mathrm{quiv}}(t_{\mathrm{col}}) + \mathbf{p}_{\mathrm{dr}}|^2 - |\mathbf{p}_{\mathrm{quiv}}(t_{\mathrm{col}})|^2]/(2\mu)$ vanishes. However, expressing $\Delta E(t_{\mathrm{col}})$ as $[|\mathbf{p}_{\mathrm{dr}}|^2 - 2\mathbf{p}_{\mathrm{quiv}}(t_{\mathrm{col}}) \cdot \mathbf{p}_{\mathrm{dr}}]/(2\mu)$, we see that if $P \gg \hbar\omega$, where $P \equiv (e^2F^2/4\mu\omega^2)$ is the cycle-average value of $|\mathbf{p}_{\mathrm{quiv}}(t)|^2/(2\mu)$ (the ponderomotive energy), $\Delta E(t_{\mathrm{col}})$ cannot vanish for most values of t_{col} unless $\mathbf{p}_{\mathrm{dr}}$ is almost perpendicular to $\mathbf{p}_{\mathrm{quiv}}(t_{\mathrm{col}})$, thereby restricting the angular range in which the photoelectron may be ejected. This reduction of phase space is the key reason for stabilization.

Actually, energy conservation need only to be satisfied to within $h/\Delta t_{\mathrm{col}}$, and therefore, writing $\Delta E(t_{\mathrm{col}}) = [d\Delta E(t_{\mathrm{col}})/dt_{\mathrm{col}}]\,\Delta t_{\mathrm{col}}$, we have $\Delta t_{\mathrm{col}} \sim \sqrt{h}/\sqrt{|d\Delta E(t_{\mathrm{col}})/dt_{\mathrm{col}}|}$. Since $d\Delta E(t_{\mathrm{col}})/dt_{\mathrm{col}} = (1/\mu)[d\mathbf{p}_{\mathrm{quiv}}(t_{\mathrm{col}})/dt_{\mathrm{col}}] \cdot \mathbf{p}_{\mathrm{dr}}$, and since $|d\mathbf{p}_{\mathrm{quiv}}(t_{\mathrm{col}})/dt_{\mathrm{col}}| \sim eF$ and $p_{\mathrm{dr}} \approx \sqrt{2\mu N\hbar\omega}$, we obtain $\Delta t_{\mathrm{col}}/T_{\mathrm{cyc}} \sim (N\hbar\omega/P)^{1/4}$. It follows that, as long as $\Delta t_{\mathrm{col}}/T_{\mathrm{cyc}} < 1$ (so that the collision is elastic), the collision duration, and hence the scattering rate, decrease as the intensity increases. The ionization rate is maximum when $\Delta t_{\mathrm{col}} \sim T_{\mathrm{cyc}}$, that is, when $P \sim N\hbar\omega$; our criterion for stabilization follows upon noting that we can put $N = 1$, since $N > 1$ implies a larger, and less probable, momentum transfer during the collision.

This simple collision picture may be used to heuristically construct the ionization amplitude [8]: The incident electron is represented by the plane-wave $|\mathbf{p}_{\mathrm{quiv}}(t)\rangle$, multiplied by the phase factor $e^{-if(t)}$, where $\hbar f(t) = \int^t dt'\,|\mathbf{p}_{\mathrm{quiv}}(t')|^2/(2\mu)$. Immediately after the collision the electron is represented by $e^{-ig(t)}|\mathbf{p}_{\mathrm{quiv}}(t) + \mathbf{p}_{\mathrm{dr}}\rangle$, where $\hbar g(t) = \int^t dt'\,|\mathbf{p}_{\mathrm{quiv}}(t') + \mathbf{p}_{\mathrm{dr}}|^2/(2\mu)$. The first-order Born amplitude for scattering from the potential W (representing the atomic nucleus), at time t_0, is $T_{\mathrm{B}}(t_0) \equiv e^{ig(t_0)-if(t_0)}\langle\mathbf{p}_{\mathrm{quiv}}(t_0) + \mathbf{p}_{\mathrm{dr}}|W|\mathbf{p}_{\mathrm{quiv}}(t_0)\rangle$, and this must be multiplied by the amplitude $\Phi(-\boldsymbol{\alpha}(t_0))$, for the electron to be at just the right position in its orbital motion to be knocked by the field into the nucleus at time t_0. Here, $\Phi(\mathbf{x})$ is the bound-state wave function, and $\boldsymbol{\alpha}(t) \equiv \int^t dt'\,\mathbf{p}_{\mathrm{quiv}}(t')/\mu$ is the displacement of the "free" electron due to the field. Averaging over the cycle time, from $t_0 = 0$ to $t_0 = 2\pi/\omega$, gives the ionization amplitude $(\omega/2\pi)\int dt_0\,T_{\mathrm{B}}(t_0)\Phi(-\boldsymbol{\alpha}(t_0))$, which is exactly the expression derived in Ref. [9] from a full quantum-mechanical analysis. Evaluating this integral by the method of stationary phase gives the energy conservation analysis discussed above at $t_0 \approx t_{\mathrm{col}}$.

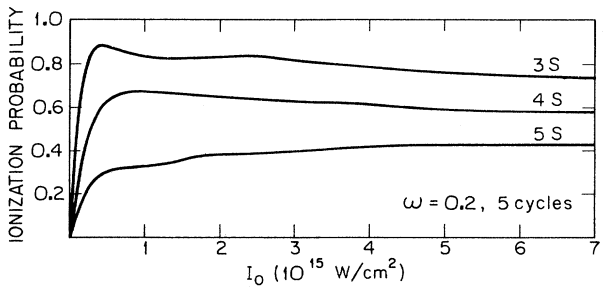

FIG. 1. Ionization probability vs the peak intensity I_0 of a linearly polarized Gaussian pulse of frequency 0.2 a.u. and a FWHM of five cycles, for atomic hydrogen initially in the $3s$, $4s$, or $5s$ state.

From Fig. 1 we see that for fixed I_0 and fixed orbital angular momentum quantum number l of the initial state the ionization probability decreases as the principal quantum number n increases; this is because the electron spends less time near the nucleus. In the perturbative regime, the ionization probability also decreases as l increases since the centrifugal barrier repels the electron from the nucleus. However, in the nonperturbative regime the mixing of the initial state with states having lower orbital angular momentum quantum number allows the electron to move closer to the nucleus, thereby enhancing the yield [10]. Furthermore, the population of the initial excited state can be transferred to less excited states which may have binding energies larger than $\hbar\omega$, and can therefore ionize rapidly by tunneling. As an illustration, we show in Fig. 2 probabilities for ionization from the $4p(m = 0)$ and $4f(m = 0)$ levels, with $\boldsymbol{\epsilon}$ the quantization axis. Again, the FWHM is five cycles, and ω=0.2 a.u. We include the $4s$ yield for comparison. The $4p(m = 0)$ and $4f(m = 0)$ ionization probabilities begin to level off, as expected, when I_0 increases toward about $I_{\mathrm{onset}} = 1 \times 10^{15}$ W/cm^2, but they begin to rise again as I_0 increases further, presumably due to coupling to states having lower orbital angular momentum quantum number (there is a three-photon resonance with the dressed $1s$ state [11], allowing rapid tunneling ionization),

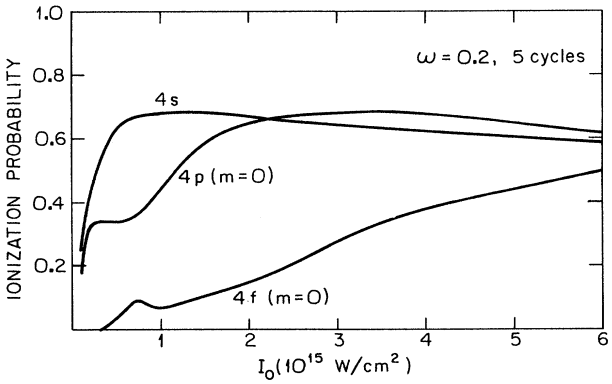

FIG. 2. Same as Fig. 1 but for the $4s$, $4p(m = 0)$, or $4f(m = 0)$ state.

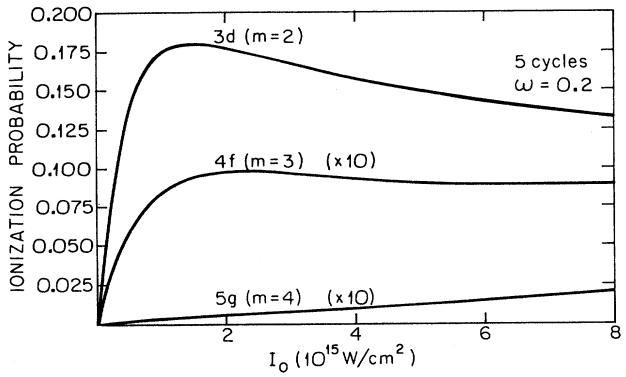

FIG. 3. Same as Fig. 1 but for the $3d(m = 2)$, $4f(m = 3)$, or $5g(m = 4)$ state.

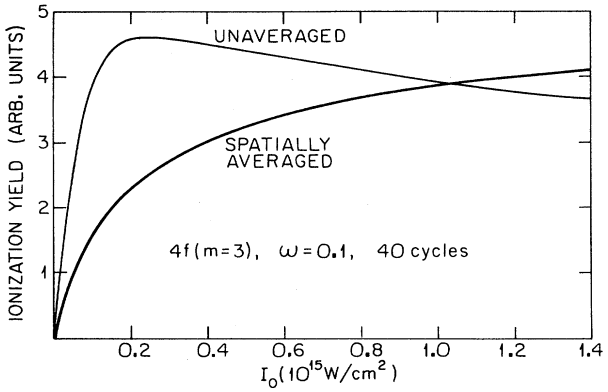

FIG. 5. Spatially averaged and unaveraged ionization yields vs I_0, for the $4f(m = 3)$ state. The frequency is 0.1 a.u. and the FWHM is 40 cycles.

though we cannot be sure this is the reason. The $4p$ state does eventually begin to stabilize when I_0 exceeds about 3×10^{15} W/cm^2, but the $4f$ state does not until much higher intensities—even a small amount of mixing with an s or a p state is apparently significant.

The initial state cannot be coupled (by linearly polarized light) to states having lower orbital angular momentum quantum number when $m = l$. Indeed, when $m = l$, the initial state is only coupled to more highly excited states, which are more stable provided that the frequency bandwidth is not too large (see below). In Fig. 3 we show probabilities for ionization of the $3d(m = 2)$, $4f(m = 3)$, and $5g(m = 4)$ states. Once again, the FWHM is five cycles, and ω=0.2 a.u. The $3d(m = 2)$ and $4f(m = 3)$ states undergo marked stabilization at the expected onset intensity, but the $5g(m = 4)$ state does not—its ionization probability continues to rise, almost linearly with I_0. However, in Fig. 4 we show the $5g(m = 4)$ ionization probabilities for the same ω, but different pulse widths. Remarkably, the $5g(m = 4)$ state does stabilize when the FWHM is ten or more cycles, and if the pulse has a peak intensity of, say, 8×10^{15} W/cm^2, fewer atoms ionize when the FWHM is 30 cycles than when the FWHM is five cycles.

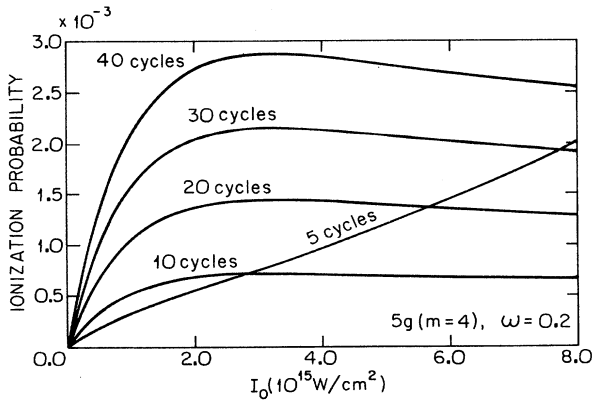

FIG. 4. Same as Fig. 1 but for the $5g(m = 4)$ state and various pulse widths.

Evidently, the frequency bandwidth $2\Delta\omega$ plays an important role, and we now comment on this. First, since $I_{\text{onset}} \propto \omega^3$, high-frequency components of the pulse cause the atom to stabilize at higher intensities. Second, energy conservation needs only to be satisfied to within the bandwidth, so "resonant" transitions to less excited (less stable) states are more likely when the bandwidth is large (though this is not possible for $m = l$ states). Third, if $\hbar\Delta\omega > |E_0|$, Raman transitions to the continuum are possible. The electron may absorb one photon and emit another of lower frequency, so as to end up with an energy slightly above the continuum threshold, and hence with a small drift momentum. We presume that absorption and emission must take place almost simultaneously—more precisely, during a time $\propto 1/\sqrt{I_0}$ — for otherwise the instantaneous momenta before and after the process would be very different due to the change in the quiver momentum [12], and then the process would involve a large, and improbable, momentum transfer. Hence, although the amplitude for two-photon Raman scattering nominally increases as I_0 with increasing I_0, the time restriction implies that the amplitude increases as $\sqrt{I_0}$, and hence the transition probability increases linearly [13]. Although still rather speculative, this may explain the (almost) linear rise of the $5s$ and $5g(m = 4)$ ionization probabilities, since for a five cycle, ω=0.2 a.u. pulse, we have $\hbar\Delta\omega$=0.017 a.u. [we define $\Delta\omega \equiv 2\sqrt{\ln(2)}/t_p = 4\ln(2)$ divided by the FWHM], while for the n=5 level $|E_0| = 0.02$ a.u.

When the bandwidth is sufficiently small, the ionization yield $Y(I_0, t_p)$ scales with t_p as $Y(I_0, t_p) = 1 - [1 - Y(I_0, t_0)]^{(t_p/t_0)}$. To prove this, we write $Y(I_0, t_p) = 1 - \exp[\int_{-\infty}^{\infty} dt\ \Gamma(I(t))/\hbar]$, where $\Gamma(I(t))/\hbar$ is the Floquet ionization rate at intensity $I(t)$; if $I(t)$ depends on t only through one dimensionless parameter t/t_p, we can scale $t \rightarrow (t_p/t_0)t$ in the integrand, which gives the scaling law after a little manipulation. We have numerically confirmed this law; as one example, the relative error in predicting the $4f(m = 3)$ ionization probability at a FWHM of 20 cycles from that at a FWHM of 10 cycles, for $\omega = 0.1$ a.u., is less than 3%, and often much less for

$I_0 < 1.2 \times 10^{15}$ W/cm^2.

In Fig. 5 we show the result of spatially averaging the $4f(m = 3)$ ionization probability for the case with a FWHM of 40 cycles and $\omega = 0.1$ a.u. We have assumed that the linear dimensions of the atomic sample are small compared to the confocal parameter and large compared to the spot size. Thus, we average over an intensity distribution $I(\rho) = I_0 e^{-2\rho^2/w_0^2}$, where w_0 is the spot size and ρ is the radial distance from the axis of the laser beam. The unaveraged yield $Y(I_0, t_p)$ exhibits stabilization at $I_{\text{onset}} = 1 \times 10^{14}$ W/cm^2, but spatial averaging masks this since the volume of atoms which experience a local peak intensity $\bar{I}_0$ below I_{onset} is very large. However, the onset of stabilization can be readily unraveled from the measured yield by differentiating with respect to I_0; the volume element dV in which the local peak intensity varies from $\bar{I}_0$ to $\bar{I}_0 + d\bar{I}_0$ is proportional to $d\bar{I}_0/\bar{I}_0$, so that $Y_{\text{expt}}(I_0, t_p) \propto \int_0^{I_0} d\bar{I}_0\, Y(\bar{I}_0, t_p)/\bar{I}_0$, and hence $I_0 dY_{\text{expt}}(I_0, t_p)/dI_0 \propto Y(I_0, t_p)$.

In conclusion, stabilization should be observable if a hydrogen atom is prepared in a high $m = l$ Rydberg state—but not too high, since we require $\hbar\Delta\omega \ll |E_0|$. As a final example, if $\omega = 0.1$ a.u. (2.7 eV), I_{onset} is only 1×10^{14} W/cm^2, and the probability for ionization of the $5g(m = 4)$ state is less than 0.06 when the FWHM is 50 cycles. We note, however, that stabilization may not occur in atoms that have loosely bound core electrons due to the possibility of core excitation or ionization by the optically driven outer electrons [14]. In addition, core electrons may tunnel out, opening other channels into which outer electrons may decay.

We thank P. H. Bucksbaum, W. E. Cooke, and B. Piraux for some helpful comments. This work was supported by the National Science Foundation, under Grant No. PHY-9017079.

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[11] Note that s states are not coupled by a three-photon transition.

[12] The change in quiver momentum is $(eF/\omega)[\cos(\omega t) - \cos(\omega t')]\epsilon$, which does not grow with increasing F if $|t - t'| \propto 1/F$.

[13] Many-photon Raman processes may be important, and can be viewed as nonadiabatic transitions to the continuum from the initial state dressed in the space-translated frame [V. C. Reed, P. L. Knight, and K. Burnett (private communication)]. In this case, the process is very nonlinear, and the condition $\hbar\Delta\omega > |E_0|$ is probably too restrictive.

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