Laser temporal and spatial effects on ionization suppression

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Ionization suppression can be defined as the decrease of photoinduced atomic ionization rates with an increased applied field intensity. The temporal and spatial intensity distribution of a focused laser pulse introduces difficulty in observing ionization suppression. An analysis of the effect of a temporally Gaussian laser pulse on ionization suppression from Rydberg levels is given. It is found that ionization suppression by the Gaussian pulse is still apparent, even though the low-intensity tails of the pulse produce moderate ionization. Further, the spatial effects are examined for a scheme whereby a narrow atomic beam is passed through the focus of a laser at which ionization is detected. Ionization suppression in the more intense region of the laser focus is predicted to be apparent above the background ionization from the relatively large region of low intensity surrounding the laser focus.

1. INTRODUCTION

Since the development of high-intensity lasers, much attention has turned toward understanding the processes governing the ionization of an atom in a strong oscillating electric field. Intuitively, one might expect the ionization rates to increase with increasing laser intensity, and this trend seems to hold over a wide variety of conditions. However, recent theoretical calculations suggest that under certain conditions an atom's ionization rate can be suppressed with increasing intensity. Ionization suppression is predicted to occur under a number of conditions. Fedorov and co-workers[1,2] showed that the one-photon ionization rates from Rydberg levels can be reduced in a strong laser field owing to an ionization-induced interference between the transitions of neighboring Rydberg levels to the continuum. Similarly, Parker and Stroud[3] have shown an inhibition of ionization owing to coherent population trapping in bound states of the atom at high intensities. Burnett *et al.*[4] have described how a stable superposition of highly excited Rydberg states can be formed that is spatially far from the nucleus. Other results by Su *et al.*[5] using a one-dimensional hydrogen model have shown that, for high enough frequencies, transitions leading to photoionization are diminished. Similar calculations by Kulander *et al.*[6] have shown that the stabilization persists also in a full three-dimensional hydrogen model. Vos and Gavrila[7] have predicted an adiabatic high-frequency stabilization (i.e., the atom becomes highly distorted, in contrast to the case of Refs. 1 and 2) in Rydberg states of high quantum number m under realistic laser temporal conditions. In recent experiments by Jones and Bucksbaum[8] and Stapelfeldt *et al.*,[9] in which barium atoms were prepared in Rydberg states, ionization of core electrons was observed without the complete ionization of the excited states when the bandwidth of the applied laser field was greater than the spacing between adjacent energy levels. This mechanism is different from that in Refs. 1 and 2, where ionization causes adjacent energy levels to broaden, overlap, and interfere, and where, to observe suppression, one must compare ionization rates for different laser intensities. Of particular interest in the present paper is the feasibility of observing ionization suppression from hydrogenic atomic Rydberg levels based on the theoretical predictions of Refs. 1 and 2 under reasonable experimental conditions. Specifically, an analysis is given of the effects of the temporal and spatial inhomogeneities of the radiation field at a laser focus.

A general difficulty associated with testing the ionization-suppression theory using a near-optical laser is the experimental constraint of a gradual turn-on and turn-off of the field. Ionization suppression is expected to occur only for strong intensities. But if a laser pulse is strong enough to permit a Rydberg level to survive owing to suppression near the pulse peak, the Rydberg level still must survive the weak portions of the pulse near its beginning and ending to allow for any observation of suppression. Nevertheless, it appears possible to surpass this difficulty with short laser pulses routinely used for laser–atom interaction experiments.[10–12] Such systems are capable of achieving subpicosecond pulses with intensities nearing 1 a.u. (3.5×10^{16} W/cm^2) or higher. A second experimental difficulty is associated with the spatial distribution of intensities at the focus of a laser beam. To achieve sufficiently high intensities to cause photoinduced ionization, one must focus the laser beam down to a small spot where it intersects a sample of atoms. Unfortunately this means that there will be atoms in both the high-intensity portion of the focus and in the much larger low-intensity portion. The difficulty lies in observing ionization suppression, which is expected to occur only in the most intense portion of the focus, above the background of ionization that is expected to occur readily in the much larger low-intensity region neighboring the focus. This difficulty may be reduced by the use of a narrow atomic beam that perpendicularly intersects a laser at its

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focus. As we will show below, the wings in space and time of focused Gaussian-shaped laser pulses do not appear to overwhelm the suppression.

2. PREDICTION OF IONIZATION SUPPRESSION

References 1 and 2 predict that, when the applied field on an atom becomes so strong that theoretical perturbative results for ionization rates become inapplicable, there exists a range of field strength for which the ionization rates from atomic Rydberg levels diminish with increasing field strength. For square pulses the one-photon time-dependent ionization probability from Rydberg level E_n ($n \gg 1$) is given by

$$w(t) = 1 - \exp(-\Gamma t), \tag{1}$$

where Γ is the ionization width. If the field is weak enough, then Γ is given by the perturbative result $\Gamma = 2\pi V^2/n^3$, where $V/n^{3/2}$ is the dipole matrix element $(1/2)\langle h|xF|E\rangle$. n is the Rydberg level, x is the usual position coordinate, F is the field strength, and E is the energy level of the final state in the continuum. Atomic units are used throughout this paper. In a semiclassical approximation, V is given by

$$V = c(FZ^{1/3}/\omega^{5/3}), \tag{2}$$

where c is the coupling constant ($c \sim 10^{-1}$ for s–p transitions[13,14]), Z is the nuclear charge, and ω is the laser frequency. The field is considered weak, and the perturbative result for Γ is applicable so long as $V < 1/\pi$. If $V > 1/\pi$, the perturbative result is no longer applicable, and, in accordance with Refs. 1 and 2, Γ is given by $\Gamma = 2/\pi^3V^2n^3$. Since V is proportional to F, it is readily seen that, in the higher field regime, Γ decreases with increasing field strength, and hence the ionization probability $w(t)$ is suppressed. This is to be contrasted with the perturbative regime, wherein Γ increases with increasing field strength.

A real laser pulse inevitably has a continuously varying field strength. Assuming a slowly and smoothly varying pulse envelope, the exponent of Eq. (1) can be replaced by an integral over time in a quasi-static approximation. Though this procedure is not rigorous, it seems to be justified if the pulse is sufficiently smooth and long (pulse duration much larger than $1/\omega$). The equation governing probability of Rydberg-level survival $(1 - w)$ in a laser pulse between times t_1 and t_2 becomes

$$P_{\text{Ryd.Sur.}} = \exp\left[-\int_{t_1}^{t_2} \Gamma(t)\mathrm{d}t\right], \tag{3}$$

where $\Gamma = 2\pi V^2/n^3$ for $V < 1/\pi$ and $\Gamma = 2/\pi^3V^2n^3$ for $V > 1/\pi$. The boundary $V = 1/\pi$, where one expression for Γ is substituted for the other, is chosen such that Γ is a continuous function of field strength. This abrupt transition between the two regimes is somewhat artificial, but the details of the transition are unknown and are assumed to be unimportant. However, since the union of the two curves produces an upward cusp, a smoothing over the transition region would tend to reduce the value of Γ relative to the model chosen, making ionization suppression more probable. In either regime the $1/n^3$ dependence shows that the higher Rydberg levels are less likely to ionize. A physical reason for this is that the higher Rydberg levels are less coupled to the nucleus and behave in a manner similar to that of free-electron states, which are unable to have any net absorption of photons in a plane wave. Returning to the definition of the parameter V of Eq. (2), we find it convenient to rewrite the parameter

$$V^2 = I/\pi^2 I_c, \tag{4}$$

where $I = F^2$ is the laser intensity that depends on the usual spatial and temporal coordinates r, z, and t, and $I_c = \omega^{10/3}/\pi^2c^2Z^{2/3}$ is the critical intensity at which $V = 1/\pi$. An estimate of I_c, with $\omega = 0.043$ a.u. ($\lambda = 1$ μm), $Z = 1$, and $c = 0.1$, is $I_c \sim 3 \times 10^{-4}$ a.u. ($\sim 10^{13}$ W/cm^2). The exact value of the constant c is not of great importance, since it can shift only the critical intensity I_c. While I_c is independent of n, it is important to recall that the formula is derived for $n \gg 1$ and that ω must be sufficiently large to ensure one-photon ionization well into the continuum. It is interesting to note that the predicted critical intensity I_c for the onset of ionization suppression is typically far below 1 a.u., in contrast with many of the other predicted forms of ionization suppression.

3. TEMPORAL EFFECTS OF A GAUSSIAN LASER PULSE ON IONIZATION SUPPRESSION

A. Calculation of Rydberg-Level Survival Probability

If a laser pulse is short enough, atoms in high Rydberg levels can survive the weak portion of the pulse, thus enabling them to experience the strong portion of the pulse, where their ionization rate is expected to be suppressed. To a good approximation, the temporal pulse envelope of short-pulsed lasers can be represented by a Gaussian function[10]:

$$I(t) = I_0 \exp\{[-(\ln 2)(2t/\tau)^2]\}, \tag{5}$$

where τ is the full width at half-maximum of the laser pulse and I_0 is the peak intensity of the pulse. Testing for ionization suppression requires that the laser pulse have a peak intensity that is well above the critical intensity I_c [see Eqs. (3) and (4)]. Using Eq. (5) in connection with Eqs. (3) and (4), we find the critical time t_c, at which $I(t_c) = I_c$, to be

$$t_c = \frac{\tau}{2}\left[\frac{\ln(I_0/I_c)}{\ln 2}\right]^{1/2}. \tag{6}$$

The integral in the exponent of Eq. (3) then separates into the intervals $[-\infty, -t_c]$, $[-t_c, t_c]$, and $[t_c, +\infty]$. With the aid of the variable substitution $u^2 = (\ln 2)(2t/\tau)^2$ and by grouping the first and last intervals together, we can write Eq. (3) as

$$P_{\text{Ryd.Sur.}} = P_1P_2, \tag{7}$$

where

$$P_1 = \exp\left\{-\frac{2\tau}{\pi(\ln 2)^{1/2}n^3}\frac{I_0}{I_c}\int_{[\ln(I_0/I_c)]^{1/2}}^{\infty} \exp(-u^2)\mathrm{d}u\right\} \tag{8}$$

and

$$P_2 = \exp\left\{-\frac{2\tau}{\pi(\ln 2)^{1/2}n^3}\frac{I_c}{I_0}\int_0^{[\ln(I_0/I_c)]^{1/2}} \exp(u^2)\mathrm{d}u\right\}. \tag{9}$$

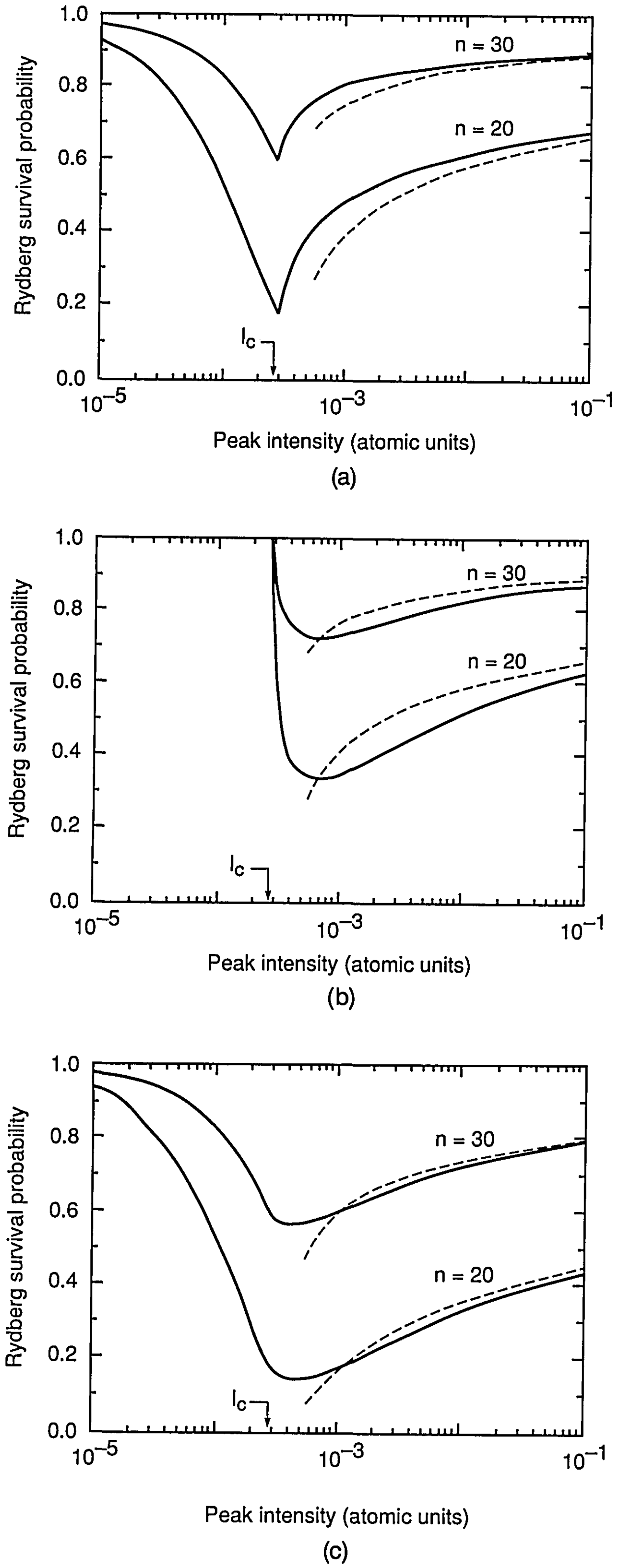


Fig. 1. (a) Rydberg-level survival probability due to the perturbative portion ($I < I_c$) of the laser pulse shown for two values of n [$P_1(I_0)$ of Eq. (8)]. (b) Rydberg-level survival probability due to the high-intensity portion ($I > I_c$) of the laser pulse shown for two values of n [$P_2(I_0)$ of Eq. (9)]. (c) Rydberg-level survival probability due to the entire laser pulse shown for two values of n [$P_1(I_0)P_2(I_0)$ of Eq. (7)]. $\tau = 2.1 \times 10^4$ a.u.; $\omega = 0.043$ a.u.; $c = 0.1$. The dashed curves are the approximations of relation (11).

The first term P_1 is associated with the portion of the laser pulse for which $V < 1/\pi$, and the second term P_2 is associated with the portion for which $V > 1/\pi$. It is this second term P_2 that is associated with ionization suppression. The integral in Eq. (8) has the form of the complementary error function, and the integral in Eq. (9) has the form of Dawson's integral. Some simplified analytical expressions can be obtained from Eqs. (8) and (9) under the assumption that the peak laser intensity I_0 is well above the critical intensity I_c. The integrals of Eqs. (8) and (9) are approximated by the expressions

$$\int_\xi^\infty \exp(-u^2)\mathrm{d}u \cong \frac{\exp(-\xi)^2}{2\xi},$$

$$\int_0^\xi \exp(u^2)\mathrm{d}u \cong \frac{\exp(\xi^2)}{2\xi} \quad \left(1 \gg \frac{1}{2\xi^2}\right), \tag{10}$$

where ξ stands for $[\ln(I_0/I_c)]^{1/2}$. Equations (8) and (9) become

$$P_1 \approx P_2 \approx \exp\left\{-\frac{\tau}{\pi(\ln 2)^{1/2}n^3[\ln(I_0/I_c)]^{1/2}}\right\}. \tag{11}$$

This approximation (valid for $I_0 \gg I_c$) shows that the contribution toward ionization from either the low-intensity or the high-intensity portion of the pulse is roughly the same. The dependence of P_1 and P_2 on peak-laser intensity I_0 calculated from relation (11) is shown in Figs. 1(a) and 1(b), respectively, by the dashed curves. The solid curves in these figures refer to the exact calculations of Eqs. (8) and (9). Physically, P_1 of Fig. 1(a) is the survival probability of the Rydberg level during the low-intensity portion of the pulse. P_2 of Fig. 1(b) is the survival probability of the Rydberg level for the high-intensity portion of the pulse. Figure 1(c) shows the Rydberg survival probability for the entire pulse P_1P_2 of Eq. (7). Again the dashed curves in Fig. 1(c) show the approximation of relation (11). Comparison of the solid and dashed curves in Fig. 1 shows that, in fact, the asymptotic expression of relation (11) provides a good approximation begin-

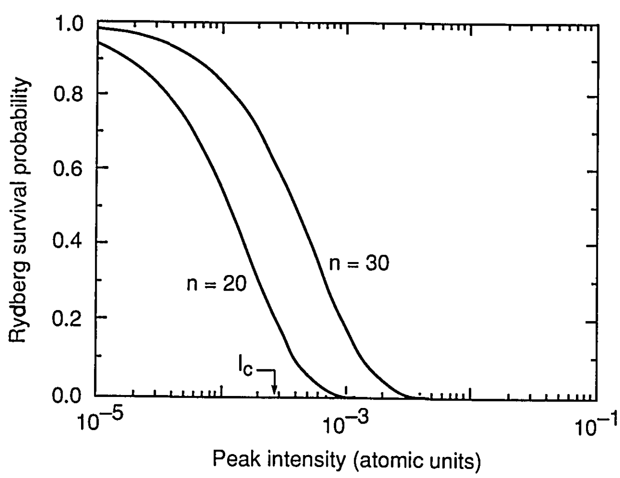


Fig. 2. Rydberg-level survival probability extending use of the low-intensity perturbative result for Γ into the high-intensity region. Perturbation theory suggests that the Rydberg survival probability should quickly tend to zero with higher intensity, in contrast with Fig. 1(c).

ing from $I_0 \sim 2I_c$. Figure 2 is given for comparison with Fig. 1(c) and shows what the Rydberg survival probability would be if use of the low-intensity perturbative result for Γ were extended into the high-intensity range ($I > I_c$) (i.e., no suppression). P_1 of Eq. (8) will produce these curves if the lower limit of the integral in the exponent is extended to zero [critical time $t_c \equiv 0$ of Eq. (6)]. In conclusion, the time dependence of the laser field does not conceal the interference suppression of ionization predicted earlier for square pulses.[1,2] The increasing parts of the curves in Fig. 1(c) correspond to the field-induced stabilization regime.

B. Estimate of the Maximum Laser-Pulse Duration Required for Suppression

In the limit where the peak intensity is much larger than the critical intensity, ($I_0/I_c \gg 1$), we can estimate the maximum pulse width required for stabilization. For survival probability to be $\gtrsim 50\%$ after the laser pulse has passed, relation (11) leads to a requirement that

$$\frac{\tau}{n^3} \lesssim \frac{\pi}{2} (\ln 2)^{3/2} \left[\ln\left(\frac{I_0}{I_c}\right) \right]^{1/2}. \tag{12}$$

4. INCLUSION OF ABOVE-THRESHOLD TRANSITIONS

An alternative expression[2] for Γ when $V > 1/\pi$ [compare with the expression following Eq. (3) above] is given by

$$\Gamma' = 2/\pi^2 V n^3. \tag{13}$$

The difference between the two formulas is as follows: Γ (where $V > 1/\pi$) of Eq. (3) is calculated on the assumption that a Rydberg electron makes one-photon transitions out of and back into the atom. If, after the interaction ceases, the electron finds itself outside the atom, then ionization occurred. The alternative expression [Γ' of Eq. (13)] incorporates the possibility that the electron absorbs more than one photon while leaving the atom, thus reducing the chance for a transition back into the atom. If one uses Γ' of Eq. (13) when $V > 1/\pi$ rather than the expression associated with Eq. (3), P_2 of Eq. (9) becomes

$$P_2' = \left\{ \exp\left[-\frac{2\tau}{\pi (\ln 2)^{1/2} n^3} \left(\frac{I_c}{I_0}\right)^{1/2} \times \int_0^{\{\ln[(I_0/I_c)^{1/2}]\}^{1/2}} \exp(u^2) \mathrm{d}u \right] \right\}^{\sqrt{2}}. \tag{14}$$

In the approximation of relations (10), $P_2' = [P_2]^2$ where P_2 is the expression given in relation (11). While the inclusion of higher-order transitions somewhat reduces ionization suppression, the Rydberg survival probability does not severely decrease.

5. SPATIAL EFFECTS ON IONIZATION SUPPRESSION IN A LASER FOCUSED ONTO AN ATOMIC BEAM

A possible way to observe ionization suppression is to pass perpendicularly a beam of atoms that are excited to known Rydberg levels through a laser focus and to detect the ionized electrons produced near the focus. It is important that the atomic beam diameter d be narrow so that the greatest possible fraction of the atoms interacts with the most intense portion of the laser beam. Since the intensity near the laser focus varies sharply with small variations in position, it is necessary to consider the cumulative effect of the focal-volume intensity distribution on the number of electrons produced. Assuming a uniform density of atoms ρ in the atomic beam, one finds the number of electrons produced at the focus by summing over all the relevant isointensity volumes Δv multiplied by their respective ionization probabilities:

$$N = \rho \sum_v [1 - P(I_v)] \Delta v = \rho \int_0^{I_0} [1 - P(I)] \left(-\frac{\mathrm{d}v}{\mathrm{d}I} \right) \mathrm{d}I, \tag{15}$$

where I_0 is the peak intensity in the laser focus. It is then important to find an expression for the volume v within an isointensity contour at the laser focus. This volume should exclude any region of the focal volume that is not intersected by the atomic beam.

High-intensity laser beams generally follow the properties of Gaussian optics, especially for long focal lengths. The intensity distribution of a Gaussian-mode laser beam near its focus is given by[15]

$$I(r, z) = \frac{I_0}{1 + z^2/z_0^2} \exp\left[\frac{-2r^2}{w_0^2 (1 + z^2/z_0^2)} \right], \tag{16}$$

where $w_0 = 2\lambda(f\text{-number})/\pi$ is the beam-waist radius, at which the intensity drops by a factor of e^2 from its peak value, and $z_0 = \pi w_0^2/\lambda$ is the Rayleigh range, defined to be the distance along the laser axis at which intensity drops by a factor of 2 from the peak. r and z are the usual cylindrical coordinates. To obtain the volume v within an isointensity contour of intensity I, one inverts Eq. (16) to give r as a function of I and z, and cylindrical disks of area πr^2 are integrated in the z direction. The limits for z are set either by the atomic beam radius $d/2$ or by the inversion of Eq. (16) for z when r is set to zero. Hence

$$v(I) = \int_{-z_{\max}(I)}^{z_{\max}(I)} \pi r^2(I, z) \mathrm{d}z, \tag{17}$$

where $z_{\max}(I, r) = \min[d/2, z_0(I_0/I - 1)^{1/2}]$. After the integration is performed, the derivative of v with respect to intensity I is found to be, with $x = I/I_0$,

$$\frac{\mathrm{d}v}{\mathrm{d}I} = -\frac{\pi w_0^2 z_0}{I_0} s(x), \tag{18}$$

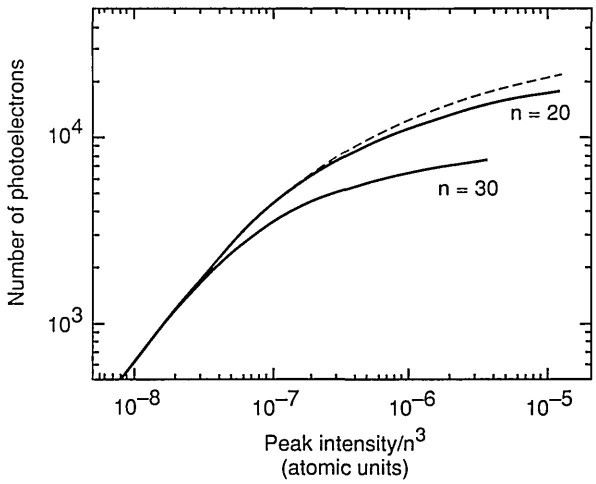
where

$$s(x) = \begin{cases} \dfrac{1}{3x^{5/2}} (1 - x)^{1/2} [1 + 2x] & \text{when } x > [1 + (d/2z_0)^2]^{-1} \\ \dfrac{1}{x} \left[\dfrac{d}{2z_0} + \dfrac{1}{3} \left(\dfrac{d}{2z_0} \right)^3 \right] & \text{when } x < [1 + (d/2z_0)^2]^{-1} \end{cases} \tag{19}$$

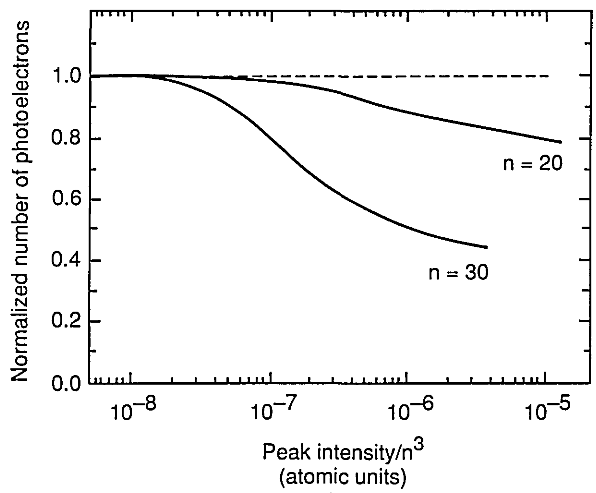
Substituting Eq. (18) into Eq. (15) gives

$$N = \rho \pi w_0^2 z_0 \int_0^1 [1 - P(x)] s(x) \mathrm{d}x. \tag{20}$$

Figure 3(a) shows the number of electrons ionized in the



(a)



(b)

Fig. 3. (a) Number of electrons produced at the laser focus plotted against I_0/n^3 from Eq. (20). The dashed curve is the n-independent perturbative result calculated always with the low-intensity expression for Γ. (b) Number of electrons produced at the laser focus normalized by the number of electrons produced in the perturbative approximation [the dashed curve of (a)]. The solid curves show the ionization-suppression result. $\tau = 2.1 \times 10^4$ a.u.; $\omega = 0.043$ a.u.; $c = 0.1$; f-number $= 20$; $d = 2$ mm.

atomic beam at the laser focus plotted against the scaled parameter I_0/n^3. The dashed curve indicates what the results would be if the perturbative Γ were used past its range of applicability to higher intensities (the same probability P is used as in Fig. 2). Since the perturbative result depends on I_0/n^3, the dashed curve is the same for any n. The solid curves are calculated with the Rydberg survival probability of Eq. (7) based on the ionization-suppression theory. The continual rise in the upper portions of the curves is due to the fact that, as the intensity of the laser increases, so does the volume in which ionization can occur. Hence, with higher laser intensity, more atoms participate in the interaction. If the number of atoms in the interaction were fixed, the curve with the perturbative result (dashed) would eventually become horizontal. The important point is that the ionization-suppression curves extend increasingly below the dashed curve, showing that they increase more slowly than the interaction volume of the laser focus. Figure 3(b) shows the same graph as Fig. 3(a), but with all curves being normalized to the dashed curve of the perturbative result. This normalization effectively factors out the volume dependence of the laser focus for the higher-intensity portion of the curves. Experimental measurement of the n-dependent curves shown in Fig. 3 would indicate ionization suppression.

The contrast of the curves in Figs. 3(a) and 3(b) can be improved by an increase in the Rayleigh range z_0 of the laser focus relative to the atomic beam diameter d. However, once the atomic beam diameter is of the order of twice the Rayleigh range of the laser, there is little to be gained in the contrast of the curves by a further increase in the laser focal length.

6. LIMITATIONS

Near the laser focus the intensity varies sharply as a function of position. However, theoretical calculations regarding ionization suppression are carried out under the assumption of a uniform plane wave. An important consideration is that the binding force on the Rydberg electron (Z^2/n^4) should be greater than the gradient of the ponderomotive potential ($\nabla I/4\omega^2$). For $\lambda = 1$ μm, this constrains n to be $\lesssim 30$ when f-number ~ 20 is used and to be $\lesssim 40$ when f-number $\gtrsim 50$ is used. In Fig. 3 the curves for $n = 40$ would appear a factor of 2 lower than those for $n = 30$. Because of experimental uncertainties in the initial n levels of the atomic beam and in the ionization measurements, it would be beneficial or perhaps essential to perform the experiment with the higher f-number in order to accommodate a higher contrast in n.

An additional consideration is the range of applicability of the ionization-suppression theory. The ionization-suppression theory[2] is expected to be valid for intensities from I_c to nI_c. For intensities above nI_c the stabilization mechanism may give way to other high-intensity processes.

7. DISCUSSION

The suppression of one-photon ionization from Rydberg levels predicted for $I_0/I_c > 1$ in Refs. 1 and 2 remains ap-

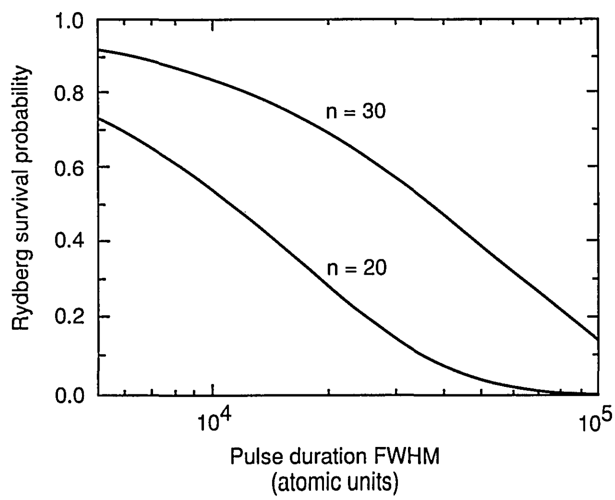


Fig. 4. Rydberg-level survival probability due to the entire laser pulse for two values of n plotted as a function of pulse duration τ [compare with Fig. 1(c)]. I_0 is set to $10I_c$. $\omega = 0.043$ a.u.; $c = 0.1$.

parent even when realistic temporal and spatial pulse shapes are included. In particular, the existence of a slow turn-on and turn-off of the laser field does not appear to cause complete ionization. A criterion for the maximum pulse duration required for observation of ionization suppression was given in relation (12). We can estimate the minimum pulse duration from the requirement that, in the absence of stabilization, the atoms within the focal volume should be completely ionized. That is, $\Gamma\tau$ must be much greater that unity, where Γ is the low-intensity perturbative result. This can be combined with relation (12) to give an approximate window of observation:

$$\frac{I_c}{I_0} \gg \frac{\tau}{n^3} < \left[\ln\left(\frac{I_0}{I_c}\right)\right]^{1/2}. \tag{21}$$

For $n = 30$ and $I_0 = 10I_c$, relation (21) becomes 3×10^3 a.u. $\ll \tau < 4 \times 10^4$ a.u. (60 fs $\ll \tau <$ 1 ps).

As one can see, the required pulse duration scales as n^3. Thus, for a given pulse duration, the window of observability can be achieved by a scaling of the choice of Rydberg level, as long as the limitations discussed in Section 6 are not disregarded. Figure 4 shows the Rydberg-level survival probability for fixed peak intensity I_0 (in this case, $I_0 = 10I_c$) plotted as a function of pulse duration τ.

The difficulty of the spatial intensity distribution near the laser focus can be overcome by the use of a narrow atomic beam intersecting the laser at the focus. The use of a high *f*-number to focus the laser beam has two advantages: it permits the atomic beam to intersect the high-intensity portion of the focus more cleanly, and it reduces the ponderomotive potential gradient associated with a tight focus.

While this paper has concentrated mainly on laser temporal and spatial effects on ionization suppression, there remain the issues associated with the experimental uncertainties in atomic beam preparation and characterization and in ionization detection. To determine the final feasibility of observing ionization suppression, one must also include detailed considerations of these factors.

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