RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 48, NUMBER 4 OCTOBER 1993

Stabilization of Rydberg atoms in superintense laser fields

Armin Scrinzi and Nils Elander
Manne Siegbahn Institute of Physics, Frescativägen 24, S-10405 Stockholm, Sweden

Bernard Piraux
Institut de Physique, Université Catholique de Louvain, 2, Chemin du Cyclotron, B-1348 Louvain-la-Neuve, Belgium
(Received 2 June 1993)

We investigate numerically the stabilization of Rydberg states of atomic hydrogen in superintense laser light. No stabilization could be observed at frequencies comparable with the Rydberg energies. We show that the stabilization observed at higher frequencies is governed by the spreading and decrease of the coupling potentials at high intensities rather than by a reduction of the wave-function amplitude in the interaction region.

PACS number(s): 32.80.Rm, 32.80.Fb

The interaction of a (one-electron) Rydberg atom with a strong electromagnetic field has recently attracted much attention, since it is under this condition that strong-field atomic stabilization is expected to be observed. By stabilization, we mean a process leading to a decreasing ionization rate with increasing field intensity, the frequency of the field being fixed. Two different physical mechanisms are at the origin of stabilization: the first one, which occurs at high frequency (photon energy higher than the ionization potential), is essentially kinematical. The second one, which occurs in a wider range of frequencies, results from an interference effect owing to Raman-type transitions via the continuum. Although these mechanisms are well understood and documented in the literature, ionization rates from very-high-lying Rydberg states have not been calculated so far [1]. In this Rapid Communication we analyze the ionization rates from Rydberg states of principal quantum number equal to 20 and discuss the above-mentioned mechanisms.

Stabilization at high frequency and high intensity is usually associated with a laser-induced delocalization, the so-called dichotomy in the atomic wave [2]. This means that the electron cloud is kept away from the nucleus such that the necessary exchange of momentum for the ejection of the electron is suppressed. A condition for the validity of this picture is a laser frequency $\omega \gg E$, where E is the binding energy of the electron in the laser field. The field intensities, on the other hand, must be such that the oscillation amplitude of a free electron in the laser field $\alpha_0 = |\mathbf{F}|/\omega^2$ at a field amplitude $\mathbf{F}$ is comparable to the atomic dimensions. This stabilization process has been studied with both time-dependent [3] and time-independent [4] methods in a case where the atom is in its ground state or a low-lying excited state. The atomic dimensions of Rydberg states $\sim n^2$ are very large. Therefore, following the established picture of high-frequency stabilization, one would expect that very large α_0 are required for stabilization. In fact, we demonstrate below that stabilization begins at much lower intensities than expected. Our results confirm those of Pont and Shakeshaft [5], who showed that stabilization starts when the ponderomotive energy becomes roughly equal to the photon energy.

A quite different mechanism for the stabilization of highly excited states was proposed, where the decay of several coherently populated Rydberg levels is inhibited by destructive interference of the transitions to the continuum [6]. This stabilization by interference was predicted to occur already at very low frequencies (comparable to, though larger than the Rydberg energies) and at intensities, where the coupling matrix element of the bound-continuum transition V_{nE} exceeds the spacing between the Rydberg levels [8]. Stabilization already at very low frequencies is what clearly distinguishes the predictions of this model from the first mechanism described above. In our numerical studies, however, we could not detect stabilization at these low frequencies.

For the experimental observability of stable configurations the time evolution of the laser intensity was shown to be important [5]. Yet the question whether stabilized states exist at all is best decided by studying the lifetime of the atom in monochromatic laser light of constant intensity. In the present work we numerically studied mechanisms of the stabilization of highly excited states in linearly polarized laser light. The observed stabilization was investigated as to whether it was of the high-frequency or of the interference type. Two laser frequencies $\omega = 0.002$ and $\omega = 0.2$ were used. (Atomic units are used unless indicated otherwise.) The larger frequency lies close to the visible, where very high laser intensities are experimentally available. The frequency $\omega = 0.002$ in the far infrared was chosen to detect possible interference stabilization in this range. Hydrogenic levels with principal quantum numbers between $n = 5$ and 20 were investigated.

All calculations were performed in the Kramers-Henneberger (accelerated) frame of reference [8], which is particularly well suited for the high-frequency regime [9]. After subtracting the ponderomotive motion and making the Floquet ansatz for the wave function, the hydrogen atom in the laser field is described by the following system of equations [9]:

$$(-\tfrac{1}{2}\Delta + V_0 - m\omega - W)\Psi_m = -\sum_{m' \ (\neq m)} V_{m-m'}\Psi_{m'}. \quad (1)$$

The potentials V_m are given by

$$V_m(\boldsymbol{\alpha}_0, \mathbf{r}) = (i^m/\pi)\int_{-1}^{1} |\mathbf{r} + \boldsymbol{\alpha}_0 u|^{-1} \times T_m(u)(1-u^2)^{-1/2}du, \quad (2)$$

where $T_m(u)$ are the Chebyshev polynomials and $\boldsymbol{\alpha}_0 = \alpha_0\hat{\mathbf{F}}$. In the limit of very high frequencies, the system of equations (1) decouples, leaving the single equation

$$(-\tfrac{1}{2}\Delta + V_0 - U)\Phi = 0. \quad (3)$$

Below we will show by explicit computation that the decoupling may already occur at quite low frequencies.

To find the quasibound states of the system we use the exterior complex scaling technique [10], where the width is found as twice the imaginary part of a complex eigenvalue $W = E - i\Gamma/2$. A mathematical proof of the applicability of the technique to the case of the hydrogen atom in a laser field can be found in [11]. The general principle of its computational implementation using finite elements basis functions is explained in [12].

For the present calculation an upgraded three-dimensional version of the program described in [12] was used. Each Floquet component Ψ_m was expanded into basis functions using polar coordinates

$$\Psi_m = \sum_{l=0}^{L} c_{ml}(\cos\theta)^l \chi_{ml}(r), \quad (4)$$

where the radial functions are expanded in a finite element basis [12] with a typical number of 200 basis functions for each χ_{ml}. The z component of angular momentum is conserved due to the cylindrical symmetry of the atom in linearly polarized light. We assume here that the atom is initially in an s state. Due to parity conservation only Floquet components with equal generalized parity $(-1)^{m+l}$ couple, which restricts the expansion by setting $c_{lm} = 0$ for $(-1)^{m+l}$ either equal to 1 or -1.

Now we briefly present the results obtained for the infrared laser frequency $\omega = 0.002$. We calculated the ionization width for three states around the $n = 20$ Rydberg level in a very restricted basis, where only s and p states ($L = 2$) with parity $(-1)^{m+l} = 1$ were included, and the Floquet blocks were restricted to one continuum block above and one block below the Rydberg states. These conditions are fairly close to the basic model assumptions made in [6] to demonstrate the stabilization mechanism by interference. According to [7], we should expect a peak of the width at the field strength where $|V_{nE}|^2 \approx |\mathbf{F}|^2/(n^3\omega^{10/3}) \approx 1/n^3$, or $\alpha_0 = \omega^{-1/3} \approx 8$. The maximum width there should be comparable to the spacing between the Rydberg levels.

Figure 1 shows the dependence of the ionization widths on α_0 in the range up to $\alpha_0 = 40$. One of the states exhibits a quite strong maximum of the width, but this maximum is not a general property of the Rydberg states, rather it is due to the one-photon resonance of

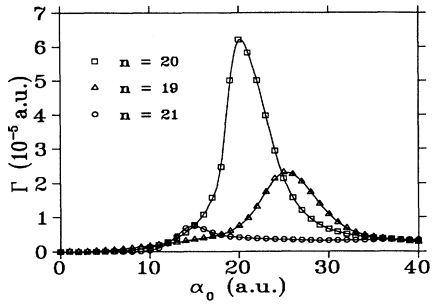

FIG. 1. Ionization widths of the Rydberg states $19s$, $20s$, and $21s$ for a laser frequency $\omega = 0.002$ a.u. The results were obtained in a strongly restricted basis including only s and p states and a total of only three Floquet blocks. The maxima are due to resonances with lower states, and disappear when these resonances are suppressed.

a lower-lying state with this particular state and it disappears when one suppresses the lower Floquet components $\Psi_{m<0}$. When on the other hand one adds more Floquet blocks, two- and three-photon resonances completely change and enhance the ionization widths. We were not able to find the proper stabilization that would show equally in all states at this very low frequency.

Another prediction of the interference mechanism is that at sufficiently high field strength the energies of the states level out at values between the original field-free Rydberg levels. Following the energy levels with increasing field we do not observe such leveling out, rather all energies keep increasing well beyond the next higher Rydberg energies.

We could not clearly identify the reason for the apparently strong disagreement between the model of interference stabilization [7] and our numerical calculations. Of course, one has to be aware of the fact that that model is formulated in terms of field-free Rydberg states, while our calculation uses the Kramers-Henneberger frame, such that the restrictions of the basis to s and p states as imposed in the present work cannot be exactly set equal to the suppression of degeneracy used in the simplest model assumptions [7]. Further, in the length or velocity gauge the bound-continuum matrix elements increase linearly with the field strength, which inevitably leads to strong coupling of the Rydberg states once the matrix elements become comparable to the spacing between the levels. Contrary to that, the bound-continuum matrix elements in the Kramers-Henneberger frame remain bounded to values that are in general much smaller than the spacing between the Rydberg levels.

Another question is whether the very low laser frequency employed here is in the range of validity of the interference model. In fact, at one step in the derivation of the basic formulas for the model [7], the approximation $\omega \approx E + \omega$ is used, which amounts to a high-frequency condition. This condition is clearly violated for our choice of parameters, which, however, lie within the ranges of validity for the model as given in [7].

Both, the model [7] and our calculation, are for constant laser intensity. We found our current results confirmed by time-dependent calculations with a realistic laser pulse shape and with parameters in the range given

above [13].

Let us now turn to the higher frequency $\omega = 0.2$. This value somewhat above the visible light was chosen to avoid having the $2p$ states in resonance with the Rydberg levels. Such resonances obscure the picture by causing spikes in the ionization rates, although the general characteristics of stabilization at higher frequencies is maintained. Again the basic mechanism of stabilization is best studied in a strongly restricted basis of s and p states, including only one Floquet block above and below the Rydberg levels.

Figure 2 shows the width of the state with principal quantum number $n = 20$ for frequencies $\omega = 0.2$ and higher. One clearly sees stabilization at high intensities. This stabilization could be identified unambiguously as being of the high-frequency type by calculating the overlap of the central Floquet component $\Psi_0(\alpha_0)$ with the corresponding solution $\Phi(\alpha_0)$ in the high-frequency limit, Eq. (3). It was found that the admixture in $\Psi_0(\alpha_0)$ of wave functions other than $\Phi(\alpha_0)$ decreases from a few percent at $\alpha_0 = 5$, where the width assumes its maximum, to less then 10^{-3} at $\alpha_0 = 15$.

We do not want to repeat here the kinematical considerations used by Pont and Shakeshaft [5] to show that effective stabilization starts when the ponderomotive energy $P = F^2/4\omega^2$ exceeds the photon energy. Instead, we would like to adopt a different point of view. The initial drop from the maximum width can be explained by a quick decay of the coupling matrix element due to the oscillating character of the continuum state. Considering for the moment the wavelength of the Rydberg wave function as long, the length scales to be compared are, on the one hand, the range of the coupling potentials V_m, which is given by the classical excursion amplitude α_0, and, on the other hand, the wavelength of the continuum state $\sqrt{1/\omega}$. From Eq. (2), one sees that the potentials V_m scale with α_0 like

$$V_m(\boldsymbol{\alpha}_0, \mathbf{r}) = \alpha_0^{-1} V_m(\hat{\boldsymbol{\alpha}}_0, \mathbf{r}/\alpha_0), \tag{5}$$

i.e., the integral of the coupling potential remains constant, but its distribution becomes flatter with increasing α_0. Once the coupling potentials spread over more than one wavelength, the coupling matrix element starts to decrease. Simple as this picture is, it allows two qualitative predictions. First, since there are only two relevant length scales, α_0 and $\sqrt{1/\omega}$, the α_0 where the width reaches a maximum scales like $\sqrt{1/\omega}$. This property is very general and also follows from the above condition on the ponderomotive energy.

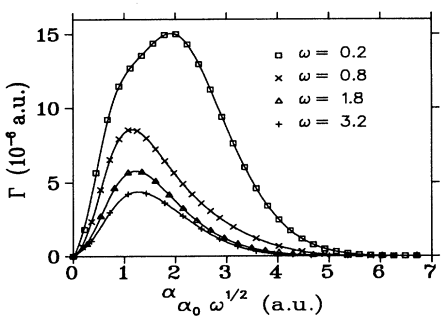

FIG. 2. Ionization widths Γ of the Rydberg state $20s$ for laser frequencies $\omega \geq 0.2$ a.u. The basis is restricted in the same manner as for Fig. 1. The decrease of the width at higher α_0 is due to high-frequency stabilization (see text). The maxima of the widths are located at $\alpha_0\omega^{1/2} \approx 1.2$.

Figure 2 shows the extent to which this scaling argument is valid for the frequencies $\omega = 0.2$, 0.8, 1.8, and 3.2. At the lowest frequency there is some deviation from the $\sqrt{\omega}$ scaling of the α_0 axis, which again is caused by the coupling to lower bound states. At the other frequencies the widths assume their maxima at $\alpha_0 \approx 1.2/\sqrt{\omega}$.

Second, the width scales with the principal quantum number n like the squares of the amplitudes, i.e., $\Gamma(\alpha_0, n) \approx n_0^3/n^3\Gamma(\alpha_0, n_0)$. This scaling behavior could be verified numerically for the α_0 around the maximum decay width (Fig. 3). For the states between $n = 10$ and 20 the scaling applies quite well, while for $n = 5$ the widths are somewhat lower than expected by the scaling law. This is quite plausible, since the maximum width is reached around $\alpha_0 = 5$, which may already significantly distort the $n = 5$ state, leading to additional stabilization. One can also directly verify that the amplitude of the wave functions in the range $|\mathbf{r}| \lesssim \alpha_0$ does not significantly depend on α_0. Over the range from $\alpha_0 = 4$ to 15 the integrals $\int_0^R |\Psi_0(\alpha_0, r)|^2 dr$, $R = 10$, 25, and 50 vary by no more than 20%, while the width decreases by several orders of magnitude.

In this picture it is the spreading of the coupling rather than the distortion of the bound state that decouples the bound state from the continuum. Of course, this spreading of the coupling potential exactly reflects the general physical reasoning that with increasing intensity there is not enough force to accelerate the electron to the one-photon continuum. The picture is also consistent with recent findings [4] that the maximal ionization widths are smaller and occur at lower α_0 than predicted in [2].

The numerical results discussed so far were obtained in a strongly restricted basis. Now we present results in a larger basis, including $L = 10$ angular momenta and five Floquet blocks $m = -2, -1, \ldots, 2$. We concentrate on the states with principal quantum number $n = 10$ and with the z component of angular momentum equal to 0. We discuss only states with parity $(-1)^{m+l} = 1$; identical conclusions can be drawn for the states with negative parity.

Figure 4 shows the widths of the states that evolve

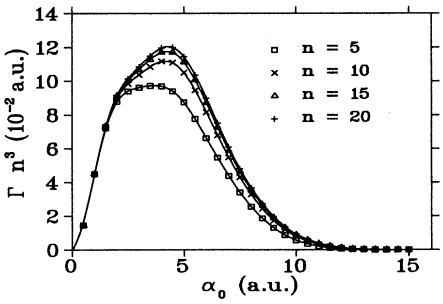

FIG. 3. Dependence of the ionization widths Γ on the principal quantum number n. Stronger deviations from the $1/n^3$ scaling are observed for the lowest state $n = 5$.

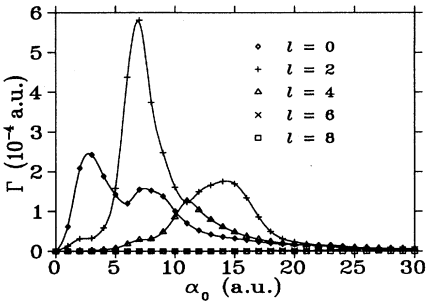

FIG. 4. Ionization widths Γ of the $n = 10$ Rydberg states that evolve from the field-free states $l = 0$, 2, 4, 6, and 8 and $l_z = 0$. The laser frequency is $\omega = 0.2$ a.u. Up to $\alpha_0 \lesssim 15$ the results are accurate on the level of $\lesssim 20\%$. The high peak around $\alpha_0 \approx 8$ is due to a 2ω resonance with the $1s$ level.

from the field-free states of angular momenta $l = 0$, 2, 4, 6, and 8. The states $l = 0$ and 2 have distinct maxima of the decay width in the range of α_0 depicted. The higher angular momentum states couple only weakly to the continuum due to the centrifugal barrier that keeps them away from the interaction region. The first maximum in the decay widths belongs to the $l = 0$ state. Surprisingly, there is a second maximum even higher than the first at larger α_0. However, this maximum is caused by the crossing of the $1s + 2\omega$ state through the region of the Rydberg states. This is reflected by an increase of the corresponding Floquet component, while the maximum disappears when one suppresses that Floquet component.

By comparing calculations with $L = 14$ and 10, both using only three Floquet blocks, we conclude that the results shown are quantitatively correct up to $\alpha_0 \lesssim 15$. Beyond that range the $l = 6$ and 8 states are expected to have higher width than given in Fig. 4. The addition of more Floquet blocks affects the widths only to the order of $\lesssim 20\%$.

The $1/n^3$ scaling law could be verified also for this basis with $L = 10$. The scaling of the α_0 axis with $\sqrt{1/\omega}$ is not as clearly visible here, since the crossing of the $1s$ state disappears for higher frequencies, while for lower frequencies crossings with the $2p$ state show up.

In summary we draw the following conclusions from our calculations. We were not able to detect the stabilization of Rydberg states predicted for very low frequencies. It may be that the interference mechanism considered for such stabilization implicitly relies on a high-frequency assumption of the type $\omega \gg E$. At higher frequencies stabilization is readily observed. For Rydberg states the stabilization is most adequately ascribed to the decrease of the coupling matrix element once the range of the interaction region in the Kramers-Henneberger frame exceeds the wavelength of the free electron. For sufficiently high Rydberg states ($n \lesssim 10$) the dependence of the decay width on α_0 around the maximum becomes independent of the principal quantum number of the state and scales simply with n^{-3}.

The authors are very grateful to R. Shakeshaft for several valuable comments on the manuscript.

[1] Ionization rates for $n \approx 7$ have been calculated recently by R. Potvliege and P.H.G Smith, Phys. Rev. A **48**, 46 (1993).
[2] M. Pont, N. Walet, M. Gavrila, and C.W. McCurdy, Phys. Rev. Lett. **61**, 930 (1988).
[3] Q. Su, J.H. Eberly, and J. Javanainen, Phys. Rev. Lett. **64**, 862 (1990); K.C. Kulander, K.J. Schafer, and J.L. Krause, *ibid.* **66**, 2601 (1991).
[4] M. Dörr, R.M. Potvliege, and R. Shakeshaft, Phys. Rev. Lett. **64**, 2003 (1990).
[5] M. Pont and R. Shakeshaft, Phys. Rev. A **44**, R4110 (1991).
[6] M.V. Fedorov and A.M. Movsesian, J. Opt. Soc. Am. B **6**, 928 (1989); **6**, 1504 (1989).
[7] M.V. Fedorov, M.Yu. Ivanov, and A.M. Movsesian, J. Phys. B **23**, 2245 (1990).
[8] W. C. Henneberger, Phys. Rev. Lett. **21**, 838 (1968).
[9] M. Gavrila and J.Z. Kaminski, Phys. Rev. Lett. **52**, 613 (1984).
[10] B. Simon, Phys. Lett. **71A**, 211 (1979).
[11] S. Graffi and K. Yajima, Commun. Math. Phys. **89**, 277 (1983).
[12] A. Scrinzi and N. Elander, J. Chem. Phys. **98**, 3866 (1993).
[13] E. Huens, B. Piraux, A. Scrinzi, and N. Elander (unpublished).