Effective Stabilization of Rydberg States at Current Laser Performances

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Stabilization, the exotic process by which an atom in a superintense laser field, of the order of the atomic unit, reacts to the increase in intensity or decrease in frequency by increasing its lifetime, has been recognized as elusive for atomic ground states. We now show that the stabilization regime can be effectively attained at present laser performances (intensities, frequencies, pulse shapes) when starting with atoms prepared initially in Rydberg states of high magnetic quantum number with respect to the axis of linear polarization.

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The behavior of atoms in superintense laser fields, of the order, or in excess, of 1 a.u. of intensity (3.5×10^{16} W/cm^2), has become a topic of lively interest and debate. On one hand, this is due to the striking features which have been predicted for this interaction regime, and, on the other, to the fact that the radiation fields required for testing them have now become available in the form of superintense, subpicosecond laser pulses, at frequencies ranging from the infrared to the ultraviolet (e.g., see Refs. [1,2]).

Some time ago the high-frequency Floquet theory of laser atom interactions developed by Gavrila and Kaminski [3–5] was applied by Pont *et al.* [6,7] to the hydrogen atom, to predict its *dichotomy* (splitting) in a monochromatic linearly polarized field of sufficiently high photon energy ω. In a subsequent work, Pont and Gavrila [8] have used the same theory to calculate ionization rates for both linearly and circularly polarized fields. It was shown by semianalytical means that in the high-frequency, high-intensity regime the atom can undergo *stabilization,* i.e., its decay by multiphoton ionization can be quenched. Whereas the idea of stabilization is a cornerstone for the validity of the high-frequency theory, it has emerged from this work in a new form. Originally it had been recognized that the ionization rates should vanish at increasing, sufficiently high ω, and fixed value of the independent parameter $\alpha_0=I^{1/2}\omega^{-2}$ a.u. [9]. It now appeared that besides this, at high frequencies and superintensities, *the decay rates decrease with I at fixed* ω, *and increase with* ω *at fixed I,* contrary to the physical understanding gained at low intensities. However, concerning the possibility of having the atom in the ground state enter the stabilization regime in a standard multiphoton ionization experiment, the conclusion was negative: The ground state cannot be stabilized at present laser performances by the adiabatic high-frequency mechanism. Similar results were found by applying the full Floquet theory by Dörr, Potvliege, Proulx, and Shakeshaft [10].

The problem of stabilization recently has been approached dynamically, by studying the evolution of wave packets according to the time-dependent Schrödinger equation. Su, Eberly, and Javanainen [11] show in the case of a one-dimensional model atom that wave-packet solutions can stabilize in intense laser pulses, i.e., will oscillate without appreciable ionization and change of shape, exhibiting at times a dichotomous structure. This *dynamical stabilization* occurs even at photon energies below the unperturbed binding energy. It has been demonstrated by Kulander, Schafer, and Krause [12] also for wave packets evolving from the ground state of real hydrogen at superintensities in the 10^{17}–10^{18} W/cm^2 range, and a (for the time being inaccessible) photon energy of $\omega=1$ a.u. Further studies on the one-dimensional model of Ref. [11], aimed at understanding the mechanism of the phenomenon and the role played by the pulse shape, were done by Burnett, Knight, Piraux, and Reed [13]. At lower intensities, where atomic distortion does not come into play, mechanisms for the suppression of ionization as the result of coherent broadband excitation of Rydberg states have also been presented (e.g., Ref. [14]).

In these dynamical studies stabilization appears as the result of the rapid turn-on of the laser pulse, which "shakes up" the atom into excited states, where it remains trapped as a consequence of their slow decay (they evolve in a quasi-high-frequency regime). As such it is a global atomic effect, quite sensitive to the shape of the pulse used, and in which the individual behavior of the atomic states in the field can hardly be distinguished. In contrast to this, the high-frequency stabilization mechanism we have studied [8] refers to *individual atomic states,* adiabatically evolving from well-defined unperturbed energy eigenstates. To distinguish it from other mechanisms, we shall call it *adiabatic stabilization.* Beyond having intrinsic significance, its interest resides in that it allows, in principle, for the possibility of eventually producing strongly distorted atoms, with states which are tunable by the intensity of the field, and long enough lived to have experiments carried out on them.

We now show that *adiabatic, high-frequency stabilization* can be achieved at present laser performances in monochromatic fields of linear polarization, starting from atomic Rydberg states of high magnetic quantum number m. (Because of the symmetry of the problem, m defined with respect to the field axis passing through the nucleus

is a good quantum number [15].) We shall specifically deal with hydrogen, although the results apply to any atom with an electron excited in a high-m state, because this problem is essentially Coulombic, too. The monochromatic case of constant intensity will be dealt with first, and that of realistic laser pulses will be addressed subsequently. We are concentrating on the case of linear polarization for which the *sufficient* condition for the validity of the high-frequency Floquet theory (see Refs. [5,6]) can be experimentally satisfied. For linear polarization this can be specialized to

$$\omega \gg |W_0^{|m|}|, \tag{1}$$

where $|W_0^{|m|}|$ is the binding energy of the *lowest-lying* state of the manifold of magnetic number m to which the initial state belongs (and not the ionization potential of the ground state, as needed in the case of arbitrary polarization). It turns out that at high frequencies ω, $W_0^{|m|}$ depends only on the α_0 defined above, and not explicitly on ω [see Eq. (3) below]. If m is chosen sufficiently high, this will decrease the unperturbed (i.e., $\alpha_0=0$) binding energy $|W_0^{|m|}(0)|$ to an extent that condition Eq. (1) can be satisfied even for infrared frequencies. For larger α_0 the condition will then be even better satisfied, as $|W_0^{|m|}(0)| > |W_0^{|m|}(\alpha_0)|$ [16].

Specifically, we have chosen to work with $m=5$, for which the lowest unperturbed state has $n=6$, and a binding energy of $|W_0^{|5|}(0)| = \frac{1}{72} = 0.0139$ a.u. This compares favorably in Eq. (1) with the photon energy of the Nd:YAlG (neodymium-doped yttrium aluminum garnet) fundamental $\omega = 1.17$ eV $= 0.0428$ a.u., and even better with that of other superintense lasers [1,2], operating at $\omega = 2.0$ eV $= 0.0735$ a.u.

To lowest order in $1/\omega$ and constant α_0 the n-photon ionization amplitude is given by [4,5]

$$f_n(\hat{\mathbf{r}}) = -(1/2\pi)\langle u_{\mathbf{k}_n}^{(-)}|V_n|u_0\rangle \quad (n \geq 1). \tag{2}$$

Here u_0 is the normalized energy eigenfunction of the initial state of the atom in the field (energy W_0), and $u_{\mathbf{k}_n}^{(-)}$ is its final, continuum eigenfunction, behaving asymptotically like a plane wave of momentum $\mathbf{k}_n$ and amplitude 1, plus an incoming spherical wave. The final energy is fixed by the energy-conservation equation $k_n^2/2 = W_0 + n\omega$. Both u_0 and $u_{\mathbf{k}_n}^{(-)}$ are solutions of the time-independent Schrödinger equation

$$[\tfrac{1}{2}\mathbf{P}^2 + V_0(\alpha_0;\mathbf{r})]u = Wu. \tag{3}$$

For linear polarization, we take the electric field of the plane wave to be $\mathbf{E} = E_0\boldsymbol{\epsilon}\cos(\omega t)$, so that the "dressed potential" V_0 in Eq. (3), as well as the V_n entering the amplitudes Eq. (2), are given by

$$V_n(\alpha_0;\mathbf{r}) = \frac{1}{2\pi}\int_0^{2\pi} e^{in\chi} V(\mathbf{r} + \alpha_0\boldsymbol{\epsilon}\cos\chi)\,d\chi, \tag{4}$$

where $V(r) = -1/r$ is the original Coulomb potential and α_0 was defined above. The differential n-photon ionization rate is

$$d\Gamma_n/d\Omega = k_n|f_n(\hat{\mathbf{r}})|^2. \tag{5}$$

In the following we shall take advantage of the high-frequency assumption of the theory [Eq. (1)] which implies that all ionized electrons (with $n=1,2,\ldots$) have large kinetic energies with respect to their initial binding energies (see the energy-conservation equation). We are then entitled to use the *Born approximation* for the final state, that is, to replace $u_{\mathbf{k}_n}^{(-)}$ by the plane wave $\exp(i\mathbf{k}_n\cdot\mathbf{r})$ [17–19].

There are two stages in the calculation of the differential rates. First comes the *computation of the initial-state eigenvalues* $W(\alpha_0)$ *and eigenfunctions* u_0 for the cases considered. Concentrating on $m=5$, we have computed the two lowest-lying states within each of the ungerade (u) and gerade (g) manifolds [15]. In order to distinguish a particular state within a manifold $\{|m|,g/u\}$ we have labeled it by the quantum numbers (n,l) of the unperturbed state to which it is connected continuously in the field-free limit, $\alpha_0=0$ (for the possibility of doing this see Ref. [7], Sec. IV); hence the notation $\{(n,l)|m|_{g/u}\}$. Thus, the states under consideration for $m=5,u$, are $\{(6,5)|5|_u\}$ and $\{(7,5)|5|_u\}$, and those for $m=5,g$, are $\{(7,6)|5|_g\}$ and $\{(8,6)|5|_g\}$ [20].

The computation of the eigenvalue problem Eq. (3) was carried out in cylindrical coordinates using the finite element method. Because of the axial symmetry the problem is two dimensional. The basis set consists of quadratic polynomials defined on triangular elements. The eigenvalues and eigenfunctions could be obtained with high accuracy by an iterative procedure developed by one of us (R.J.V.) [21]. In Fig. 1, we present the "correlation diagrams" of the levels, i.e., their α_0 dependence starting from the unperturbed Rydberg atom limit ($\alpha_0=0$), to dichotomous atom limit at large α_0. The energy of the state $\{(6,5)|5|_u\}$ is the lowest of the whole $|m|=5$ manifold (u or g), and, as mentioned already, its energy should be taken as a reference for the applicability of the theory to ionization from any of the states of the manifold. The decrease with α_0 of the binding energies in Fig. 1 is monotonic, but quite slow in comparison to that of the ground state (see Ref. [7], Fig. 1). There are no avoided crossings. By inspection of the eigenfunctions we have ascertained that dichotomy sets in, in all cases, for α_0 between 500 and 1000.

The second stage of the computation consists in the *evaluation of the integral in Eq. (2),* with the u_0 calculated. This reduces to a time-consuming threefold numerical integration [over two space coordinates and the parameter χ in Eq. (4)]. Angular integration of the differential cross section Eq. (5) yields the n-photon ionization rate Γ_n, and summation over all n the total rate Γ of the state u_0. It turns out numerically that *at these high values of m, for all* α_0, $\Gamma_n \ll \Gamma_1$ $(n>1)$, so that $\Gamma \cong \Gamma_1$ (as is the case for any state to lowest order in perturbation theory with respect to I or α_0). Indeed, we

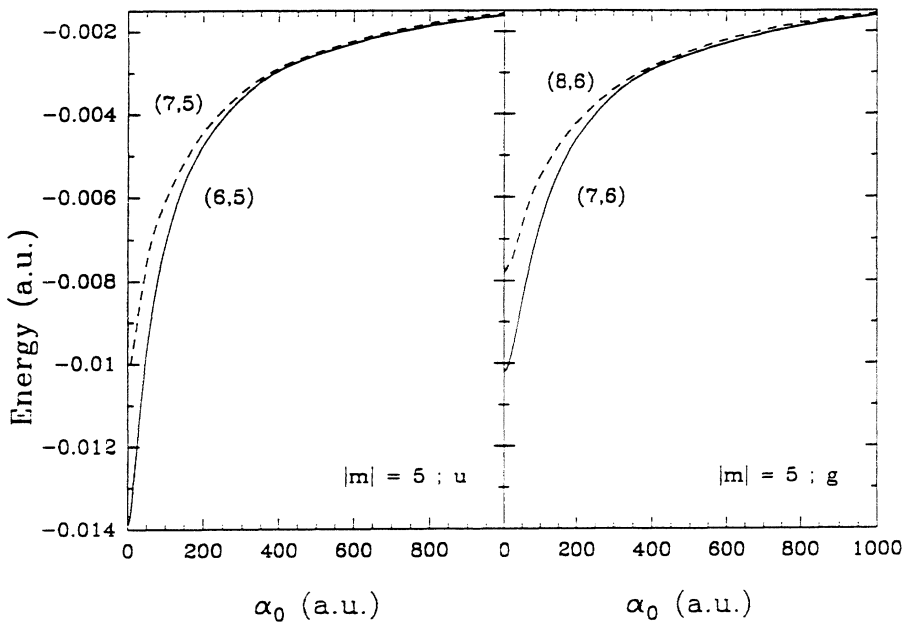

FIG. 1. Energies (in a.u.) of the two lowest-lying states of the symmetry manifolds $\{|m|=5,u\}$ and $\{|m|=5,g\}$ of atomic hydrogen, for linear polarization, as a function of α_0 (in a.u.). The states are designated by the quantum numbers (n,l) of their $\alpha_0=0$ limits.

could confirm analytically that for a given m, the Γ_n decrease roughly as $n^{-|m|-2}$ $(n \gtrsim 1)$. In Fig. 2 we show the lifetime $\tau=1/\Gamma$ of the state $\{(7,6)|5|_g\}$ as a function of I, for two frequencies. The curves display a similar behavior as those for the ground state [8]. At smaller intensities, τ has the typical I^{-1} variation required by perturbation theory, and then, after passing a minimum, has a (m-dependent) power-law increase with I, typical of the stabilization regime. In this regime τ is larger for the lower frequency. As for the ground state, stabilization starts at much lower α_0 than dichotomy. The essential difference from the ground state is, however, that now the minimal lifetime is much longer. For

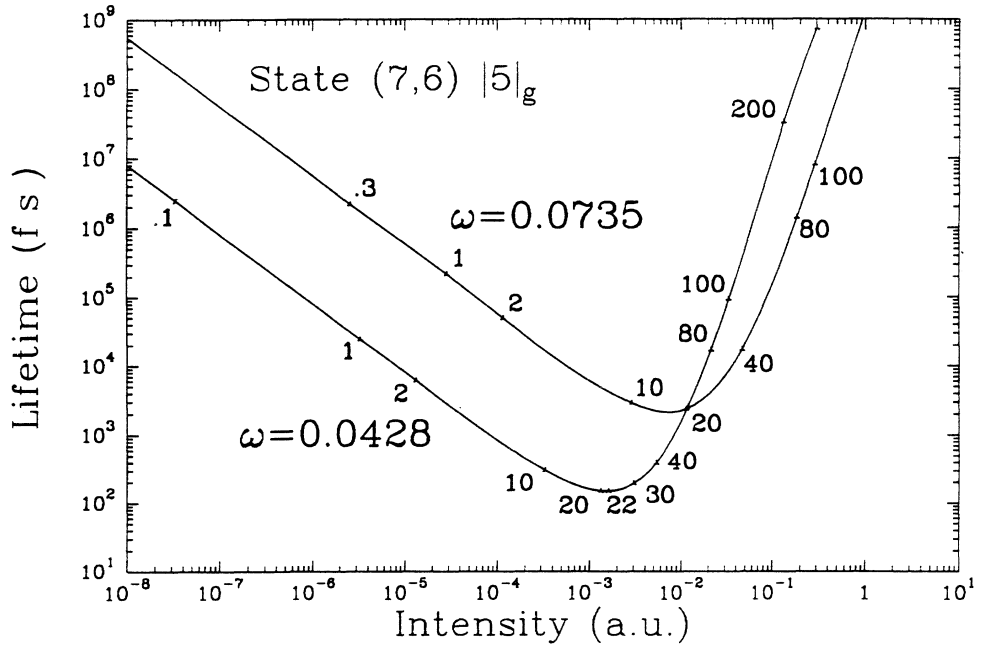

FIG. 2. Lifetime (in fs) of hydrogen in the state $\{(n=7,l=6),m=5,g\}$ for linear polarization, as a function of the intensity (in a.u.), at $\omega=1.17$ eV=0.0428 a.u., and $\omega=2.0$ eV=0.0735 a.u. The numbers adjacent to the points on the curves give the corresponding values of α_0 (in a.u.). Note that the minimum lifetime on the $\omega=0.0735$ curve is $\tau=2000$ fs (at $\alpha_0=15.5$), i.e., very much longer than the 100-fs laser pulses available at this photon energy (see Ref. [2]).

$\omega = 2.0$ eV $= 0.0735$ a.u. (see Refs. [1,2]), we find a *minimal lifetime of 200 fs.* Moreover, stabilization begins now at lower intensities (below 0.1 a.u.), and the increase of τ with I is much steeper.

From the constant intensity case considered above we now turn to the physical situation of an atom initially prepared in a well-defined $(n,l,m=5)$ state, irradiated by a subpicosecond laser pulse. We can allow *a posteriori* the intensity I to become time dependent if (a) the variation of the field amplitude is sufficiently small during a light period, so that the notion of frequency remains meaningful, and bandwidth effects do not influence the dynamics; (b) the variation of the intensity is sufficiently slow on the atomic time scale, so that sudden "shake-up" transitions to higher discrete states or the continuum, other than those due to multiphoton ionization, can be suppressed, i.e., the atom remains in its initial, adiabatically changing state [22]. Condition (a) is satisfied in all cases of practical interest. An analysis based on the adiabatic perturbation theory applied to the Hamiltonian in Eq. (3), with a realistic pulse shape, shows that condition (b) also can be met for the states shown in Fig. 2, if the pulse duration is not shorter than about 100 fs [23]. Moreover, for pulse durations of up to several hundred femtoseconds, *the atomic lifetime amply exceeds the pulse duration* for the cases shown in Fig. 2, so that practically all atoms exposed will survive the turn-on of the pulse and will enter the stabilization regime.

We thus want to conclude that adiabatic stabilization of individual, high-m atomic states can be achieved with present laser capabilities. Because of its very characteristic features, its signature on ionization should be possible to detect. One such experimental scheme will be discussed elsewhere.

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[15] The classification of the atomic states into symmetry manifolds was discussed for linear polarization in Ref. [7].

[16] See Fig. 1.

[17] The fact that the electron energies $k_n^2/2$ are large with respect to $|W_0^{|m|}|$ (and not necessarily with respect to the ground-state binding energy $|W_0^{|0|}|$), should be sufficient to insure the applicability of the Born approximation to the continuum solutions of Eq. (3) in the subspace of magnetic quantum number m, and it is only the component of $u_{\mathbf{k}_n}^{(-)}$ lying in this subspace which is relevant for Eq. (2).

[18] Although the Born approximation represents a substantial simplification of the computation of Eq. (2), it may limit the validity of the result to higher values of ω than required by condition Eq. (1).

[19] Equations (2) and (3) of the high-frequency theory describe ionization in the oscillating, Kramers-Henneberger frame of reference, see Refs. [4–7]. A plane wave in this frame corresponds to a Gordon-Volkov wave in the laboratory frame.

[20] We have switched here to this numeral notation in order to avoid possible confusion arising from the literal notation adopted in Ref. [7] when applied to highly excited states.

[21] Details of the method will be published elsewhere.

[22] When already one-photon ionization is possible [as with Eq. (1)], and the energy curves $W(\alpha_0)$ show no avoided crossings (as is the case with the lowest u and g states in Fig. 1), resonances and diabatic transitions are of no concern [e.g., see R. Potvliege and R. Shakeshaft, Phys. Rev. A **38**, 4597 (1988)].

[23] When dealing with higher-m states the turn-on time of the field should be lengthened in order to be able to maintain adiabaticity.