Localization of Floquet States in the rf Excitation of Rydberg Atoms

R. Blümel and U. Smilansky[a]
Max Planck Institute for Quantum Optics, D-8046 Garching, Federal Republic of Germany
(Received 28 July 1986)

We study the Floquet states which determine the response of Rydberg atoms to an external rf field. We discuss the recently observed transition from narrow to broad states and show that the expansion amplitudes of the bound-space–projected Floquet states on high-n bound states of the unperturbed atom decay as a power of n. Therefore, even the broad states are marginally localizable. We also derive an expression for the critical value of n where the transition between narrow and broad states occurs and use this relation to interpret recent measurements of rf ionization of high-n states in hydrogen.

PACS numbers: 31.60.+b, 05.45.+b, 32.80.Rm

Radiation-induced excitation and ionization of Rydberg atoms is presently the subject of a very intensive experimental[1-5] and theoretical[5-11] effort. Of central importance for the quantum description of this process is the determination of the nature of the Floquet eigenstates.[12] In particular, their degree of localization or delocalization determines the way by which energy is transferred to the atom. In the localized situation the induced excitation is limited, whereas if delocalized Floquet states overlap with the initial state of the atom, energy can be transferred efficiently until the atom finally ionizes. Compared with the kicked rotor[13-15] the analysis of the Floquet states of the periodically perturbed hydrogen atom is complicated by the presence of the ionization continuum: The Floquet states are generally not normalizable. However, recent research work[7-11] indicates that the initial stages of the ionization process take place within the bound space of the atom with little participation of the continuum states. Therefore, a careful study of the bound-space component of the Floquet states is necessary for a deeper understanding of the ionization mechanism. On the basis of semiclassical arguments and some numerical data,[5,6] it was recently suggested that among the bound-space–projected Floquet states there exist both localized and "delocalized" states. Confining our attention to the dynamics of the one-dimensional model of Rydberg atoms,[5-11] we show that for a given driving field the Floquet eigenstates can be divided into two major categories: (a) states which overlap mainly with low-n bound states and which differ very little from the unperturbed atomic states (narrow states) and (b) states which overlap mainly with high-n states and whose expansion amplitudes on the bound states of the atom decay as a power of n for large n (broad states). The exponent can take the value $-\frac{5}{2}$ or $-\frac{3}{2}$ depending on the way one treats the coupling to the continnum. Because of the power law we characterize the broad Floquet states as being "marginally localizable" within the bound space of the atom. The division of the Floquet states between the two classes sometimes leaves a finite number of intermediate states which do not correspond exactly with either class. There is, however, an important domain of field parameters where the transition between the two classes is abrupt. This domain is the one relevant to the recent ionization experiments[1-3] and will be the main subject of the present discussion.

We shall derive an approximate expression for the critical value n_t where the transition between the two regimes (a) and (b) occurs, and we will demonstrate its significance for the explanation of recent experiments of Koch and collaborators.[2,3]

First, we shall consider the dynamics in the space spanned by the bound eigenstates of the free atom. This is the quantum analog of the classical treatment of diffusion in the action-angle phase space. The connection between classical diffusion and quantum localization should be made on the basis of this model which will be referred to as model B (B for bound). The effects of the coupling to the ionization channel will be studied by investigating two approximate and complementary methods. In the first (method C), we retain the continuous nature of the positive-energy spectrum at the expense of neglecting continuum-continuum (CC) coupling.[10,16] In the second approximation (CC) we use a Sturm representation,[7] where the interaction is fully accounted for, but the continuum is discretized.

An objective measure for the extension of the Floquet states $|\Psi^{(\alpha)}\rangle$, when expanded in the basis $|n\rangle$, when expanded in the basis $|n\rangle$ of the bound atom, is the width function $W(n)$[17]:

$$W(n) = \exp\left[-\sum_{\alpha} |\langle n|\Psi^{(\alpha)}\rangle|^2 \ln(|\langle n|\Psi^{(\alpha)}\rangle|^2)\right]. \quad (1)$$

It is assumed that $\sum_\alpha |\langle n|\Psi^{(\alpha)}\rangle|^2 = 1$. $W(n)$ gives the effective number of states used in the expansion of the state $|n\rangle$ in the Floquet basis. We obtain the Floquet eigenstates by solving the Schrödinger equation

$$i|\dot{\phi}(t)\rangle = [H_0 + V(t)]|\phi(t)\rangle \quad (2)$$

subject to the boundary conditions

$$|\phi^{(\alpha)}(T)\rangle = e^{-\omega_\alpha}|\phi^{(\alpha)}(0)\rangle, \tag{3}$$

where $T=2\pi/\omega$. The Floquet states are the eigenstates of the one-cycle propagator, and hence $|\Psi^{(\alpha)}\rangle \equiv |\phi^{(\alpha)}(0)\rangle$. The corresponding quasienergies are ω_α. Addressing the dynamics in the bound space (model B), we have

$$(H_0)_{nm} = -(1/2n^2)\delta_{nm}; \quad V(t) = \epsilon \sin(\omega t) X. \tag{4}$$

Atomic units are used throughout and X is the position operator.

The operators H_0 and X differ in their dependence on n. While the eigenvalues of H_0 *decrease* rapidly with n, the expectation values $\langle n|X|n\rangle$ *increase* quadratically with n. Hence the Floquet eigenstates will display a transition from being close to H_0 eigenstates at low n to approaching the eigenstates of X in the high-n domain. This picture is clearly supported by the behavior of two typical width functions displayed in Fig. 1. If we extract explicitly the large diagonal part $V_D(t)$ of the interaction, the critical value of n where the transition occurs can be estimated by an appropriate perturbation expansion which is carried out in the interaction representation:

$$|\phi^{(\alpha)}(t)\rangle = \exp\left[-i\left(H_0 t + \int_0^t V_D(t')dt'\right)\right]|\phi_I^{(\alpha)}(t)\rangle. \tag{5}$$

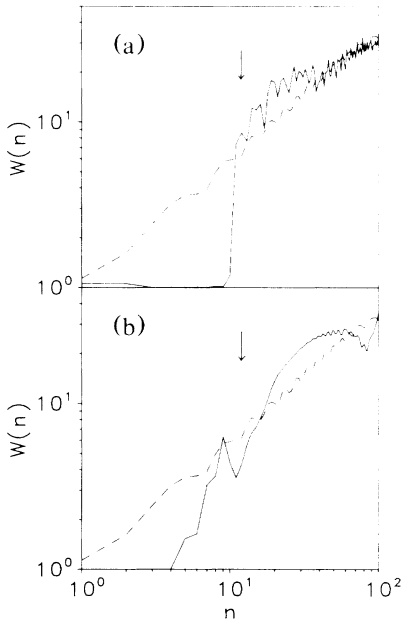

FIG. 1. Solid line: Width $W(n)$ of the unperturbed states $|n\rangle$ in the quasienergy basis. (a) $\epsilon=10^{-5}$, $\omega=5\times10^{-5}$ ($\omega n_t^3<1$), (b) $\epsilon=5\times10^{-5}$, $\omega=5\times10^{-3}$ ($\omega n_t^3>1$). Dashed line: Width of the unperturbed states $|n\rangle$ in the eigenbasis of the bound-space–projected X operator. The arrows indicate the transition point calculated according to Eq. (8).

Substituting (5) into Eq. (2), integrating over a field cycle, and using Eq. (3), we obtain

$$\left\{\exp\left[i\left(H_0 T + \int_0^T V_D(t')dt'\right) - i\omega_\alpha\right] - 1\right\}|\phi_I^{(\alpha)}(0)\rangle = \frac{1}{i}\int_0^T \tilde{V}_I(t')\,|\phi_I^{(\alpha)}(t')\rangle dt', \tag{6}$$

where $\tilde{V}_I(t)$ is the interaction representation of the nondiagonal part of $V(t)$.

In the low-n regime we start the perturbation expansion by setting $|\phi_I^{(\alpha)}(t)\rangle = |n\rangle$. Then $\omega_\alpha = -(1/2n^2)T$ and the Floquet state $|\Psi^{(n)}\rangle$ which is centered on $|n\rangle$ is distributed on $m\neq n$ states according to

$$|\langle m|\Psi^{(n)}\rangle| = \left|\frac{2\epsilon X_{nm}}{\omega_{z_{nm}}}\sum_{s=1}^{\infty}(-i)^s\frac{s^2}{s^2-\Delta_{nm}^2}J_s(z_{nm})\right|, \quad \Delta_{nm}=\frac{1}{2\omega}\left(\frac{1}{n^2}-\frac{1}{m^2}\right); \quad z_{nm}=\frac{3\epsilon}{2\omega}(m^2-n^2). \tag{7}$$

The Floquet state $|\Psi^{(n)}\rangle$ is localized on $|n\rangle$ as long as the amplitudes (7) are small. Except for resonance conditions, the largest amplitudes occur for $m=n\pm1$. According to Hose and Taylor,[18] the critical n at which localization breaks down is estimated as the value $n=n_t$ for which $|\langle n_t+1|\Psi^{(n_t)}\rangle| = \frac{1}{2}$. Using the leading terms in Eq. (7) and $X_{n,n+1}\approx -\frac{1}{3}n^2$, we get

$$\frac{1}{2} = \left|\frac{\frac{1}{3}\epsilon\omega n_t^8}{1-(\omega n_t^3)^2}\left(1 - i\frac{1-(\omega n_t^3)^2}{1-(2\omega n_t^3)^2}\frac{3\epsilon n_t}{\omega}\right)\right| \tag{8}$$

which, away from resonances, reproduces the transition points n_t to better than ±5 states. This is illustrated in Fig. 1 by the arrows which mark the value n_t calculated from Eq. (8). We used Eq. (8) to calculate n_t for the parameters used in Fig. 1 of Ref. 8. The resulting $n_t=73$ agrees remarkably well with the n value where the probability distribution in this figure exhibits the sharp transition.

Given a value of n, one can use Eq. (8) to calculate the critical field ϵ_c for which the transition to broad states occurs at the given n. The full line in Fig. 2 shows the critical field evaluated from Eq. (8). The stars were obtained by calculation of numerical the width function. The analytical estimate of the critical field is less accurate in the vicinity of the one- and two-photon resonances at $n=87$ and $n=69$, respectively. This is expected on the basis of the crude criterion chosen to determine n_t.

We observed that the transition between localized and broad states occurs abruptly when $\omega n_t^3<1$ [Fig. 1(a)]

and more gradually when $\omega n_t^3 > 1$ [Fig. 1(b)]. The different rates of transition have important consequences for the ionization process, which could be tested experimentally. It should also be noted that various resonances or avoided crossings of Floquet eigenvalue[10,11] may introduce deviations from the simple picture described above.

Directing our attention to the Floquet states which have a large overlap with high-n states $|n\rangle$, we expect their bound space component to resemble the eigenstates $|\xi_N\rangle$ of the bound-space-projected X operator, which have the following properties: (1) The eigenvalues ξ_N are discrete and for large N behave like $\xi_N \simeq (\pi^2/8)(N+\frac{1}{4})^2$. (2) The eigenstates are normalizable. Their expansion amplitudes on the eigenstates $|n\rangle$ of H_0 are given approximately by

$$\langle n|\xi_N\rangle \simeq \begin{cases} 0 & \text{if } n < \frac{1}{4}\pi(N+\frac{1}{4}), \\ \text{const}\times n^{-5/2} & \text{if } n > \frac{1}{4}\pi(N+\frac{1}{4}). \end{cases} \quad (9)$$

(3) In the $n > \frac{1}{4}\pi(N+\frac{1}{4})$ region the $n^{-5/2}$ behavior is modulated by a finite number of oscillations. The last zero of $\langle n|\xi_N\rangle$ occurs at $n = (\pi/4\sqrt{6})(N+\frac{1}{4})^{3/2}$.

These results were obtained by semiclassical arguments and were checked numerically by a direct diagonalization of the X operator in a basis of 1000 bound states. At this point we can repeat the perturbative calculation of the Floquet eigenstates choosing a representation where the operator X is diagonal. Assuming an X eigenstate to represent the Floquet state to zeroth order, one finds that the mixing of neighboring X eigenstates due to the presence of H_0 is $\sim 1/\sqrt{N}$ for large enough N values. This substantiates the claim that the Floquet states which overlap appreciably with high-n states approach in the limit the $|\xi_N\rangle$ states. This is demonstrated in Fig. 1 where the width functions are compared with the width of the n states in the X eigenbasis (dashed line). Because of the power-law structure of the $|\xi_N\rangle$ states [Eq. (9)] the width function should depend linearly on n for large n. Apart from basis truncation effects, which become serious for $n > 70$, this dependence is indeed observed [see, e.g., Fig. 1(a)].

The addition of continuum effects (method C) does not change the picture by much. The probability to remain in the bound space decreases as a result of ionization, but solving Eq. (2) with the initial condition $n(t=0) = \delta_{nn_0}$ shows that for large enough n, $\langle n|\phi(t)\rangle \approx a(t)n^{-5/2}$, where $a(t)$ is independent of n, proving again the $n^{-5/2}$ localization pattern.

To investigate the role of continuum-continuum coupling we introduce method CC where again one may obtain the Floquet eigenstates numerically and project them on the H_0 eigenstates $|n\rangle$. The only difference between the present and the former methods is the power law—the X eigenstates decay now as $n^{-3/2}$, and the decay of the Floquet states follows the same power-law decay. However, this change does not affect the values obtained for n_t which are the same for the three methods.

To show that the $n^{-3/2}$ power law is not an artifact of the asymptotic decay properties of the chosen representation, we diagonalized the X operator which corresponds to our model C in the Sturmian basis. The eigenstates of this X operator reproduce the $n^{-5/2}$ power law. The eigenstates of the complete X operator show the $n^{-3/2}$ behavior.

Because of the power-law decay, there is a significant difference between the structure of the Floquet eigenstates discussed here and in the case of the periodically kicked rotor[13-15] where the Floquet states are either exponentially localized[15] or nonnormalizable extended states[14] with a corresponding continuous spectrum.

In previous studies[10,11] we have shown that for ionization to occur, probability has to be transported to a specific range of bound states $|n\rangle$, the "ionization window" states, which are characterized by $n > n_W = (1/3\epsilon)^{1/4}$. If the atom is initially in a state $n_0 < n_t(\epsilon, \omega) < n_W$, the atom will be only weakly ionized because there are no Floquet states which have large overlap with both the initial state and the window states. Once n_0 exceeds n_t there exist Floquet states which extend to the window domain with amplitudes which decrease slowly (power law) with n. Therefore, given the value of ω and the initial state n_0, we can use (8) to determine the critical field strength ϵ_c for which the onset of strong ionization is expected to occur. In this way we analyzed recent experiments[2,3] where the critical ionization field was measured. Figure 2 shows the experimental data together with our theoretical results. The continuous line is the solution of Eq. (8). Away from the resonances and for $n < 60$ it reproduces the numerical and experimental data. The stars show the values of the critical field obtained from a numerical evaluation of the propagator (method CC) and finding the field strength for which

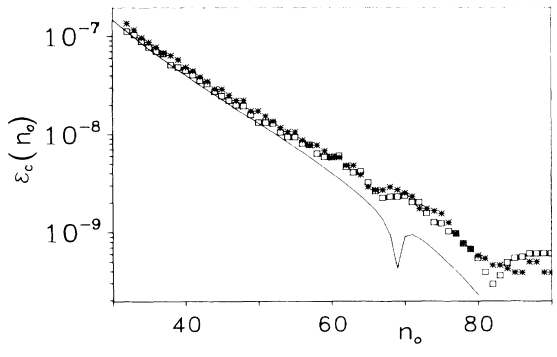

FIG. 2. The critical ionization field $\epsilon_c(n_0)$ as a function of the initial state n_0 for frequency $\omega = 1.5\times10^{-6}$ a.u. Squares: experimental results (Refs. 2 and 3). Asterisks: numerical calculation of the field at which $n_t = n_0$. Solid line: perturbative approach [Eq. (8)].

$n_t = n_0$. The agreement between the calculations and the data is very good in the region $n_0 < 75$ where the numerical determination of the transition point is unique. At higher n values the transition point becomes ambiguous because of the proximity of the resonance and the more gentle rise of $W(n)$ at n_t, which characterizes the region $\omega n_t^3 > 1$.

We would like to thank Professor P. M. Koch for communicating his data prior to publication and for stimulating discussions. One of us (U.S.) is grateful to Professor H. H. Walther for the hospitality at the Max Planck Institute for Quantum Optics. This research was supported in part by the Israeli Basic Research Commission.

---

[a]Permanent address: Department of Nuclear Physics, The Weizmann Institute, 76 100 Rehovat, Israel.

[1]J. E. Bayfield and P. M. Koch, Phys. Rev. Lett. **33**, 258 (1974); J. E. Bayfield, L. D. Gardner, and P. M. Koch, Phys. Rev. Lett. **39**, 76 (1977).

[2]K. A. H. van Leeuwen, G. v. Oppen, S. Renwick, J. B. Bowlin, P. M. Koch, R. V. Jensen, O. Rath, D. Richards, and J. G. Leopold, Phys. Rev. Lett. **55**, 2231 (1985).

[3]P. M. Koch, private communication; P. M. Koch *et al.*, in "The Physics of Phase Space," edited by W. W. Zachary (Springer, Berlin, in press).

[4]J. N. Bardsley and B. Sundaram, Phys. Rev. A **32**, 689 (1985).

[5]J. N. Bardsley, B. Sundaram, L. A. Pinnaduwage, and J. E. Bayfield, Phys. Rev. Lett. **56**, 1007 (1986).

[6]G. Casati, B. V. Chirikov, and D. L. Shepelyansky, Phys. Rev. Lett. **53**, 2525 (1984).

[7]G. Casati, B. V. Chirikov, D. L. Shepelyansky, and I. Guarneri, Phys. Rep. (to be published).

[8]G. Casati, B. V. Chirikov, I. Guarneri, and D. L. Shepelyansky, Phys. Rev. Lett. **56**, 2437 (1986).

[9]N. B. Delone, B. P. Krainov, and D. L. Shepelyansky, Usp. Fiz. Nauk **140**, 355 (1983) [Sov. Phys. Usp. **26**, 551 (1983)].

[10]R. Blümel and U. Smilansky, Z. Phys. D (to be published).

[11]R. Blümel and U. Smilansky, Phys. Scr. **35**, 15 (1987).

[12]Ya. B. Zeldovich, Zh. Eksp. Teor. Fiz. **51**, 1492 (1967) [Sov. Phys. JETP **24**, 1006 (1967)].

[13]G. Casati, B. V. Chirikov, F. M. Izraelev, and J. Ford, in *Stochastic Behavior in Classical and Quantum Hamiltonian Systems,* Lecture Notes in Physics Vol. 93 (Springer, New York, 1979), p. 334.

[14]F. M. Izrailev and D. L. Shepelyansky, Theor. Math. Phys. **43**, 553 (1980), and Dokl. Akad. Nauk SSSR **249**, 1103 (1979) [Sov. Phys. Dokl. **24**, 996 (1979)].

[15]S. Fishman, D. R. Grempel, and R. E. Prange, Phys. Rev. Lett. **49**, 509 (1982); D. R. Grempel, R. E. Prange, and S. Fishman, Phys. Rev. A **29**, 1639 (1984).

[16]R. Blümel and U. Smilansky, Phys. Rev. A **32**, 1900 (1985).

[17]R. Blümel and U. Smilansky, Phys. Rev. Lett. **52**, 137 (1984).

[18]G. Hose and H. S. Taylor, Phys. Rev. Lett. **51**, 947 (1983).