Physica Scripta. Vol. 35, 15–19, 1987.

Microwave Ionization of Hydrogen Atoms Below the Classical Chaos Border

R. Blümel

Dept. of Theor. Physics, TU München, 8046 Garching, FRG

and

U. Smilansky*

Max-Planck-Inst. for Quantum Optics, 8046 Garching, FRG

Received September 1, 1986; accepted September 7, 1986

Abstract

We present and discuss theoretical predictions for the occurrence of radiation induced ionization of hydrogen atoms in fields which are well below the classical ionization threshold. Strong ionization occurs due to enhanced population of a band of high n states which ionize easily. This enhancement happens only at rather narrowly defined field values, and is explained in terms of avoided crossings of Floquet levels.

An enormous experimental [1–4] and theoretical [4–9] effort is currently concentrated on a system which one might call the "standard system" of Quantum Chaos: A highly excited hydrogen atom in a spacially homogeneous monochromatic microwave field. The study of this quantum problem – especially its ionization dynamics – is of great interest because it involves a simple quantum system whose classical counterpart shows a clear transition from regular (no ionization) to chaotic motion (onset of ionization) [8] and for which a rich body of experimental data is already available. Moreover it offers the opportunity to study an atom in an external electromagnetic field which is very strong compared to the atomic binding field. Thus, the rf-driven hydrogen atom might very well be regarded as a first step toward the more general many-electron laser-atom interaction problem. From the practical point of view there is yet another reason to choose the rf-driven hydrogen atom as the standard system of Quantum Chaos: It is possible to prepare the highly excited atoms in extremal Stark states [10], e.g., with parabolic quantum numbers $(n, n-1, 0)$. For large principal quantum numbers n these states are extremely elongated in the positive z direction with negligible extension in x- and y-direction [11]. It can be shown [9] that they are approximately eigenstates of the 1-dimensional Hamiltonian [12, 13]

$$H_0 = \begin{cases} \frac{p^2}{2} - \frac{1}{z} & z > 0 \\ \infty & z \leqslant 0 \end{cases} \tag{1}$$

where p and z are momentum and position of the atomic electron in z-direction. Atomic units are used in eq. (1) and throughout this paper. Since the different states within a given Stark manifold are only weakly coupled by the applied microwave field [9], the external perturbation induces mainly transitions in the principal quantum number n and the atom stays 1-dimensional even in the presence of the external time-varying field. To a very good approximation the dynamics of the highly excited hydrogen atom is therefore described by [2]:

$$H(z, t) = H_0 + \varepsilon \cdot z \cdot \sin(\omega t) \tag{2}$$

where ε and ω are the field strength and field frequency respectively. It has to be noted however that according to recent 3-dimensional classical calculations [14] the elongated electron orbits show a tendency to precess, and a small static field has to be added to the Hamiltonian (2) in order to fix the orbits' orientation [4, 10]. But even in the presence of a static field component (which will not be considered here) the possibility of a 1-dimensional description dramatically reduces the amount of technical labor in the theoretical description of this periodically driven quantum system.

The probability to ionize a Hydrogen atom by a monochromatic radiation field shows a characteristic dependence on the field strength [1–4]. At sufficiently low values of the field the ionization is vanishingly small and it rises abruptly at a critical value of the field to which we shall refer as the ionization threshold. The threshold field ε_c depends on the field frequency ω, the initial state $|n_0\rangle$ in which the atom is prepared and on the time the atom is allowed to interact with the external driving field. In most cases the ionization probability $P_i(\varepsilon)$ is a monotonic function of the field strength ε, rising quickly at $\varepsilon = \varepsilon_c$, which permits an experimental extraction of the critical ionization field within reasonable limits. The analysis of the ionization experiments was so far carried out within the framework of classical mechanics and a very good overall agreement with the data has been obtained [4]. However, some ionization curves deviate drastically from the simple behavior described above, and non monotonic dependence on the field strength was observed (see, e.g., Fig. 7 of Ref. [3]). Some of these structures occur close to or even below the classical ionization threshold where sealing KAM-curves do not allow any ionization. Obviously we are here in the presence of a new and unexpected quantum phenomenon [15] not anticipated by previous classical work on the ionization dynamics of the driven hydrogen atom (see, e.g., Ref. [8]) for a review). We propose a mechanism to explain the "subthreshold" ionization bumps and we will demonstrate their purely quantum origin.

Our quantum mechanical treatment of the ionization problem will be based on the 1-dimensional Hamiltonian (2).

* Permanent address: Dept. of Nucl. Physics, The Weizmann Institute, 76100 Rehovot, Israel.

The unperturbed Hamiltonian H_0 possesses a discrete and a continuous spectrum

$$H_0|n\rangle = -\frac{1}{2n^2}|n\rangle; \quad H_0|k\rangle = \tfrac{1}{2}k^2|k\rangle. \tag{3}$$

We define the projection operators $\hat{P} = \Sigma_n|n\rangle\langle n|$ and $\hat{C} = \int dk|k\rangle\langle k|$ which project on the bound and continuum spaces respectively. Multiplying the Schrödinger equation

$$i|\dot{\psi}\rangle = H(z, t)|\psi\rangle \tag{4}$$

from the left by $\hat{P}$ or by $\hat{C}$ we arrive at the following set of coupled equations for the P-space (C-space) part of the wave function $|\psi\rangle$:

$$i|\dot{\psi}_P\rangle = H_{PP}|\psi_P\rangle + H_{PC}|\psi_C\rangle \tag{5a}$$

$$i|\dot{\psi}_C\rangle = H_{CP}|\psi_P\rangle + H_{CC}|\psi_C\rangle \tag{5b}$$

An obvious shorthand notation was used for the projected wave functions and operators. Since the probability to ionize an atom at time t is given by:

$$P_I(t) = |\langle\psi_C(t)|\psi_C(t)\rangle|^2 = 1 - |\langle\psi_P(t)|\psi_P(t)\rangle|^2 \tag{6}$$

it is enough to know the P-space dynamics of the atom, which is also interesting for direct comparison with classical ideas about a diffusive growth of energy in the P-space. The continuum is eliminated from eq. (5) by formally integrating eq. (5b). Introducing the C-space Greens function $G_C(t, t')$ we can write:

$$|\psi_C(t)\rangle = \frac{1}{i}\int_0^t G_C(t, t')H_{CP}(t')|\psi_P(t')\rangle\, dt' \tag{7}$$

Inserting this result into eq. (5a), we arrive at an equation for the P-space part of the wave function which is completely equivalent to the original Schrödinger equation (4) and of course as difficult to solve:

$$i|\dot{\psi}_P(t)\rangle = H_{PP}|\psi_P(t)\rangle + \frac{1}{i}\int_0^t H_{PC}(t) \times G_C(t, t')H_{CP}(t')|\psi_P(t')\rangle\, dt'. \tag{8}$$

The equation becomes more tractable if we neglect continuum-continuum coupling (ccc). In this case the continuum Greens function reduces to $G_C(t, t') = \exp(-i(t - t')(H_0)_{CC})$. To substantiate the validity of this approximation we performed a few additional checks. A careful study [16] of the response of an electron subjected simultaneously to an attractive $\delta(z)$-force and an external periodic perturbation shows that even for fields much stronger than will be considered here, ccc has nearly no effect on the ionization rates.

In another test we calculated the 1-photon ionization rate for $n_0 = 30$ and $\omega_0 = 30$ as a function of $\varepsilon_0 = \varepsilon n_0^4$. We reproduce by our method the 1-photon ionization results obtained in Ref. [6] where a Sturm basis was used. But since the results of this test are also reproduced by an analytical formula [17] obtained from first order perturbation theory, this test checks the consistency of our numerical procedure rather than the influence of ccc. On the other hand it indicates that if we keep the ionization rate small, or in other words, if we use the ionization channel only as a weak probe on the P-space dynamics, our approximate treatment of the continuum is adequate. We are aware of the recent observation of "above threshold ionization" [18] where ccc seems to play a major role. Still, neglecting ccc is a very reasonable approximation as long as we are interested in the integrated ionization rate and not in the detailed momentum distrubution of the ejected electrons. More detailed information on the method of integrating eq. (8) can be found in Ref. [19] and in a forthcoming publication [20].

If we want to understand the mechanisms by which a hydrogen atom ionizes, we have to find the states which are most efficiently coupled to the continuum. As a matter of fact only a limited number of states provide the link between the P-space and the continuum. The existence of this "ionization window" is easily demonstrated in a model which consists of a single bound state level $|n\rangle$ coupled to the continuum by the external microwave field. It was checked numerically that after an initial transient which does not last for more than one cycle, the occupation probability of such a level decays exponentially in time and the resulting decay rate per cycle

$$I_R = \frac{2\pi}{\omega}\dot{P}_I(t)/P_I(t) \tag{9}$$

serves as a convenient measure to characterize the importance of such a level as a doorway to the continuum. Figure 1 shows the single state decay rates which result from such a numerical experiment for various values of the applied field ε and $\omega = 4 \times 10^{-4}$. Recently we were able to obtain an analytical expression [20] for the single state ionization rate $I_R(n; \varepsilon, \omega)$ defined above. It reproduces the numerical data very well, and it has the remarkable scaling property:

$$I_R(n; \varepsilon, \omega) = \frac{\varepsilon^{13/12}}{\omega}\tilde{I}_R(\varepsilon n^4) \tag{10}$$

$\tilde{I}_R$ is independent of ω. It increases abruptly at $\varepsilon n^4 = 1/3$ and decays as $(\varepsilon n^4)^{-1}$ for large arguments. Thus, the ionization window starts at $n_W^{(1)} = (1/3\varepsilon)^{1/4}$, and consists of all the states $n \leqslant n_W^{(2)}$ for which $\tilde{I}_R(\varepsilon n^4)/(\tilde{I}_R)_{max}$ is larger than, say, 1/10.

It is important to note that for all the experiments conducted so far [1–4], $n_0 < n_W^{(1)}$ is always fulfilled since the "window condition" corresponds to a scaled field of $\varepsilon_0^{(W)} = \varepsilon n_W^{(1)4} = 1/3$, which is approximately three times larger than the scaled field $\varepsilon_0^{(st)} = \varepsilon^{(st)} n_0^4 \approx 0.12$ needed for static field ionization. The starting state $|n_0\rangle$ is therefore always essentially decoupled from the continuum and in order to achieve appreciable ionization in fields weaker than $\varepsilon_0 \approx 1/3$, probability has to be transferred from the starting state $|n_0\rangle$ to the

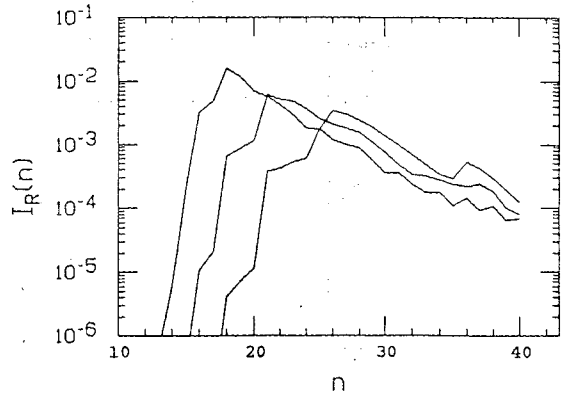

Fig. 1. Ionization rate $I_R(n)$ per field cycle for a single state n coupled to the ionization continuum for field strength $\varepsilon = 4.48 \times 10^{-6}$, 2.24×10^{-6} and 1.12×10^{-6}. The field frequency is $\omega = 4 \times 10^{-4}$.

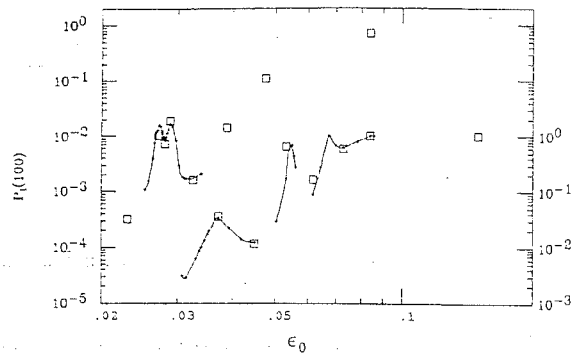

Fig. 2. Ionization probability of the state $|n_0 = 10\rangle$ after 100 field cycles as a function of the scaled field strength $\varepsilon_0 = \varepsilon n_0^4$. Upper data for $\omega_0 = \omega n_0^3 = 0.429976$ (left scale), lower data for $\omega_0 = 0.39926$ (right scale). Squares: full numerical solution of eq. (8). "Cheap" calculations [20] (diamonds) interpolate between the results obtained from the full calculations. The lines through the symbols serve only to guide the eye.

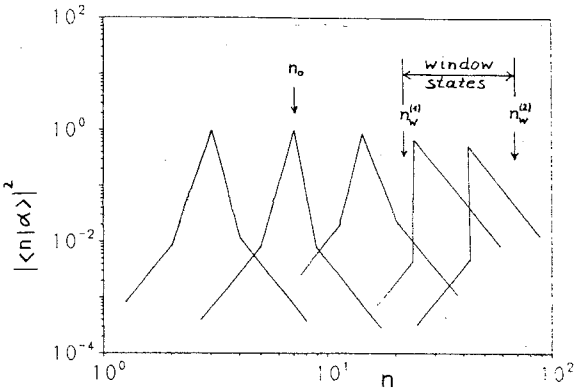

Fig. 3. Schematic sketch of the overlaps $|\langle n|\alpha\rangle|^2$ of some typical Floquet states in a weak field condition ($\varepsilon n_0^4 \ll 0.1$).

window states with $n_W^{(1)} \leqslant n \leqslant n_W^{(2)}$. The ionization process, as we see it, is therefore a two step process which consists of an excitation stage which feeds the window, followed by a subsequent transfer of probability from the ionization window states to the continuum.

The subthreshold ionization resonances [3] correspond to a situation where the field is weak and in the general case unable to excite the window states. At first sight there seems to be no way to produce appreciable ionization in a case where the occupation probability shows no tendency to move from its starting state $|n_0\rangle$ where this state by itself is dynamically decoupled from the continuum (see Fig. 1 and remember $n_0 < n_W^{(1)}$). However, a numerical solution of the time dependent equations (8) for $n_0 = 10$, $\omega_0 \approx 0.43$ and $\omega_0 \approx 0.40$ as a function of the field strength clearly shows subthreshold ionization peaks, which rise well above the average ionization signal (see Fig. 2). In both cases we checked that the peaks occur at field strengths for which sealing KAM-trajectories do not allow any upward diffusion in action. Classical dynamics prohibits ionization in these cases and the name "subthreshold resonances" is therefore justified.

In order to gain insight into the mechanism which induces subthreshold ionization, we decouple the continuum altogether and restrict our further investigations to the P-space dynamics alone. Here it is useful to discuss the problem in terms of the one-cycle propagator – or "Floquet operator" – U which carries the system over one complete cycle of the applied microwave field:

$$|\psi(t + T)\rangle = U(T)|\psi(t)\rangle, \quad T = 2\pi/\omega. \tag{11}$$

The spectral properties of the Floquet operator will turn out to be of crucial importance to explain the excitation mechanism responsible for the subthreshold peaks. We diagonalize the one-cycle propagator:

$$U(T)|\alpha\rangle = e^{i\omega_\alpha}|\alpha\rangle \tag{12}$$

The Floquet operator U is a unitary operator. The quantities ω_α, the "quasi-energies" [21], are therefore real numbers corresponding to the eigenstates $|\alpha\rangle$ of U, which are called the "quasi energy states". Once the Floquet operator is calculated, the time evolution of the wave function can be described by a quantum mapping [22] and the amplitude to excite any state $|n\rangle$ after N cycles of the microwave field and for the initial condition $|n_0\rangle$ is given by:

$$\langle n|\psi(NT)\rangle = \langle n|U^N(T)|n_0\rangle = \sum_\alpha \langle n|\alpha\rangle\, e^{i N\omega_\alpha}\langle\alpha|n_0\rangle \tag{13}$$

Hence, in order to enable a $n_0 \to n$ transition, there has to be at least one quasi energy state in the sum (13) which connects both $|n\rangle$ and $|n_0\rangle$. In other words, for ionization to occur, we need a relatively broad quasi energy state which has a large overlap with the initial state and at least one of the window states.

Figure 3 shows schematically some relevant Floquet states in a typical weak field condition (compare also Fig. 1 in Ref. [7]). The quasi energy states which have a large overlap with states in the vicinity of $|n_0\rangle$ are extremely narrow, and although the states are getting broader toward the window, none of them connects $|n_0\rangle$ to the window domain. This confirms pictorially what we know already: For weak fields we cannot excite the window states and there will be no ionization.

However, if we look at the quasi energies ω_α as a function of the applied field, we see that a subthreshold peak is always accompanied by an avoided crossing of quasi energies. Figure 4 shows the subthreshold peaks of Fig. 2 on a more refined scale together with the corresponding crossings of the quasi energies. The avoided crossing of quasi energies provides us with the excitation mechanism which we were looking for: Assume that before the crossing of two quasi energies ω_α and ω_β, the corresponding quasi energy states have the following properties:

$$|\langle n_0|\alpha\rangle| = A \sim 1; \quad |\langle m|\beta\rangle| = B \sim 1;$$
$$|\langle n_0|\beta\rangle| \sim 0; \quad |\langle m|\alpha\rangle| \sim 0, \tag{14}$$

where as before $|n_0\rangle$ stands for the state in which the atom was initially prepared and $|m\rangle$ is some window state, efficiently coupled to the continuum. At the point of avoided crossing, the quasi energy eigenstates are denoted by $|\alpha^{(+)}\rangle$ and $|\alpha^{(-)}\rangle$ and they will be essentially odd and even linear combinations of the states $|\alpha\rangle$ and $|\beta\rangle$:

$$|\alpha^{(\pm)}\rangle \approx \frac{1}{\sqrt{2}}(|\alpha\rangle \pm |\beta\rangle) \tag{15}$$

Both, $|\alpha^{(+)}\rangle$ as well as $|\alpha^{(-)}\rangle$ will now efficiently connect the

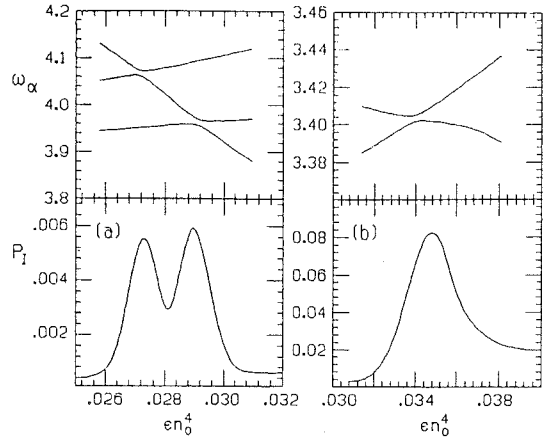

Fig. 4. Ionization probability as a function of the driving field strength in the vicinity of the lowest subthreshold resonance. (a) $P_1(N = 100)$, $\omega_0 = 0.429976$. (b) $P_1(N = 300)$, $\omega_0 = 0.39926$. The relevant Floquet levels are plotted on top.

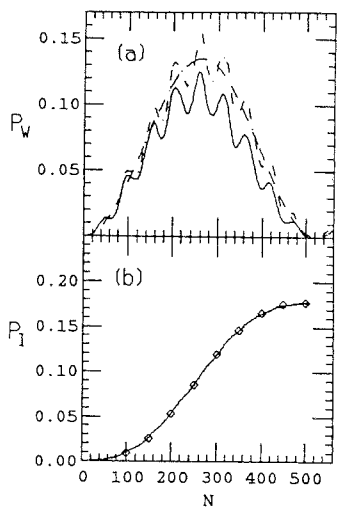

Fig. 5. (a) The dependence of the window probability on the number of cycles N. continuous line: full calculation including coupling to the continuum. Dash dot line: continuum decoupled. Broken line: the contribution from the two crossing Floquet states [eq. (16)]. (b) The dependence of the ionization probability on the number of field cycles N. Continuous line: full calculation. Diamonds: time integrated P_W renormalized as explained in the text. Field parameters: $\omega_0 = 0.429976$, $\varepsilon_0 = 0.0271$.

state $|n_0\rangle$ to the ionization window and a large enhancement of the $n_0 \to m$ transition followed by a measurable amount of ionization from the state $|m\rangle$ is expected. At the field which corresponds to the point of avoided crossing, one expects the transition probability to behave as:

$$P_{n_0 \to m}(N) = |\langle m|U^N(T)|n_0\rangle|^2 \approx |A \cdot B|^2 \times \sin^2\left(\frac{\omega_\alpha - \omega_\beta}{2} N\right). \tag{16}$$

In Fig. 5(a) we show the total occupation probability of the ionization window

$$P_W(N) = \sum_{m=n_W^{(1)}}^{n_W^{(2)}} |\langle m|\psi(NT)\rangle|^2 \tag{17}$$

for the resonance condition $\varepsilon n_0^4 = 0.0271$, $\omega n_0^3 \approx 0.43$. The full line is $P_W(N)$ which was obtained from a numerical solution of eq. (8). It is compared with a continuum decoupled calculation (dash–dotted line) and the time structure agrees very well but for the slow reduction due to ionization. The dashed line is the prediction of eq. (16) with $\omega_\alpha - \omega_\beta = 0.006$. This line reproduces the slow N-dependence of $P_W(N)$ but misses the higher frequency component which is due to the interference of the nearby Floquet state which also has a large overlap with the $n_0 = 10$ state. In Fig. 5(b) we compare the ionization probability $P_1(N)$ (full line) with the time integral of $P_W(N)$ (diamonds). The complete matching was obtained after $\int P_W(N)\, dN$ was multiplied by a mean value of the ionization rate ($\bar{I}_R = 3 \times 10^{-3}$) in the window domain.

These results substantiate the qualitative validity of our interpretation of the sub-threshold ionization resonances as being due to an enhanced excitation of the "window" states followed by ionization.

A few remarks are now in order:

(a) The search for subthreshold ionization and a detailed study of the time structure of the ionization probability probes the level-crossing mechanism, which, in a sense, is a generalization of the resonance concept to the strong field, multi-level regime.

(b) The distance between quasi energies could be extracted experimentally from the time differentiated ionization signal $P_1'(N) = \mathrm{d}/\mathrm{d}N\, P_1(N)$. According to the above results we have

$$P_1'(N) \sim P_W(N) \sim \sin^2\left(\frac{\omega_\alpha - \omega_\beta}{2} N\right).$$

The distance of two crossing quasi energies is therefore given by $|\omega_\alpha - \omega_\beta| = 2\pi/\Delta N$ where ΔN is the separation between consecutive maxima of $P_1'(N)$ ("Quasi energy spectroscopy").

(c) The occurrence of Floquet crossings and its drastic influence on the relevant dynamics is of general nature and it occurs in other systems [23, 24]. It is a genuine quantum effect whose importance may be less apparent the more semi-classical the system is. One needs a detailed calculation to be able to locate avoided crossings of relevant Floquet levels, and this is why they are difficult to predict. In particular one cannot expect that the values of the parameters for which crossing occurs are related by scaling laws to other crossing points. This contrasts with the apparent success of classical scaling when applied to the gross features in the ionization threshold data [4]. In spite of this, avoided crossings of quasi energies offer a very selective means to induce desired transitions and this might find some important applications.

At the same time that we proposed theoretically the existance and explanation for the occurrance of subthreshold resonances, the Stony Brook group found experimental evidence in the form of irregularities and bumps in the measured ionization signals [3]. The most prominent structure occurs for $n_0 = 38$ as a clear peak which cannot be reproduced by the classical calculations [25]. In Fig. 6, we show the results of our preliminary quantum calculations together with the preliminary experimental data. The most significant point to observe is that the calculations predict rather accurately the positions and the widths of the ionization maximum and minimum for the $n_0 = 38$ case. The $n_0 = 37$ maximum also fits nicely the preliminary experimental observations [26]. In these calculations we establish that the rate for subthreshold

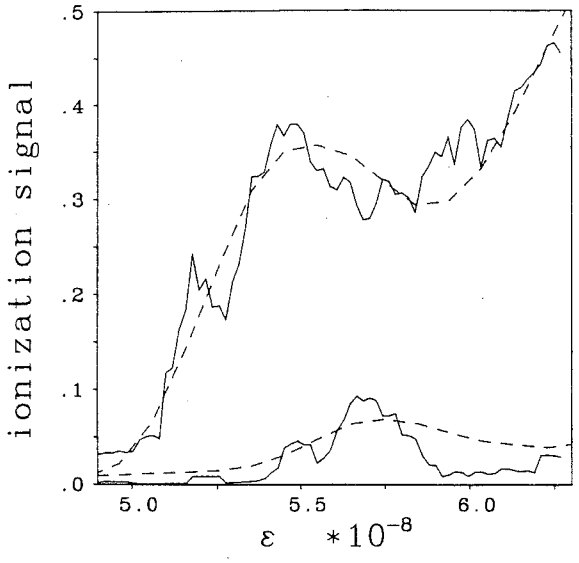

Fig. 6. Ionization signals for $n_0 = 38$ (upper curves) and $n_0 = 37$ (lower curves) as a function of the microwave field strength ε. The frequency is fixed at $\omega = 1.5085 \times 10^{-6}$. The theoretical signals (200 microwave cycles, full lines) were averaged as explained in the text. The $n_0 = 38$ ($n_0 = 37$) signal was reduced by a factor 1.8 (4.0) to match the experimental data [26] (≈ 300 cycles including adiabatic switching of the field, broken lines).

ionization is determined by the ability to excite the "window" states. Working within the bound space, we checked that the mechanism responsible for the excitation of the window states is indeed the avoided crossing of quasi energy states discussed above. In the present case there is an accumulation of several important crossings at the position of the observed peak, whose combined effects produce the broad structure of the ionization peak.

The coupling to the continuum was carried out by solving a simplified version of eq. (8), in which the memory kernel matrix was replaced by a time local, diagonal matrix. This procedure amounts to adding an imaginary part to the diagonal coupling matrix elements, whose strength was determined by the simple single state ionization rates [eq. (10)]. We cannot expect that this simple model reproduces the absolute value of the ionization signal, but on the other hand it contains the main physics – the avoided crossing of the (now complex) quasi energies. The crude method of coupling the continuum is responsible for the fact that the theoretical ionization signal is overestimated. We reduced the calculated signals by a factor of 1.8 and 4.0 (for the $n_0 = 38$ and $n_0 = 37$ cases respectively) to match the experimental data.

The individual crossings of levels produce sharp ionization spikes (with a field width of $\approx 2 \times 10^{-11}$ a.u.). The experimental accuracy is however approximately 1% in the field strength and $\approx 0.5\%$ accuracy in frequency. The theoretical results shown in Fig. 6 were obtained after averaging the calculated signal over $\Delta\varepsilon = 4 \times 10^{-10}$ a.u. intervals.

Summarizing our results, we have identified a new quantum mechanism which explains recently observed ionization peaks below the classical chaos border. This mechanism is based on the occurrence of an avoided crossing of quasi energies of the corresponding Floquet operator. We suggest to measure the separation of the crossing quasi energies experimentally by a careful analysis of the time dependent ionization probability of a hydrogen atom in a relatively weak external microwave field.

Acknowledgement

We would like to thank Prof. P. M. Koch for communicating the experimental results of his group prior to their publication. One of us (U. S.) would like to thank Prof. H. Walther for the hospitality at the MPQ in Garching.

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