# Transitions induced by separatrix crossing

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We discuss the role of separatrix crossing in a classical Hamiltonian system for a description of transitions in the corresponding quantum system. We develop a simple picture of resonant trapping of classical states and its semiclassical analog. Its relevance is illustrated by numerical analysis and by pointing out its verification in microwave excitation of highly excited hydrogen.

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## I. INTRODUCTION

Recent experimental and theoretical research has corroborated the long-standing idea that the description of quantum-mechanical processes may be simplified by invoking generic features of the corresponding classical motions; regular and stochastic flows are, of course, the key notions.

Unfortunately, theoretical research had to be confined to systems with but a few degrees of freedom. More precisely, the majority of all investigations focused on systems (like hydrogen atoms in strong magnetic fields) with two essential degrees of freedom, or, even simpler, on periodically driven one-dimensional systems with "1.5 degrees of freedom." It is the latter that we shall adhere to in this paper.

Before going into specifics, however, we first sketch in a rather cursory manner the central points of the line of arguments employed in setting up a quasiclassical description of quantum transitions induced by the variation of external parameters, the problem we shall focus on.

Suppose we are given a Hamiltonian  $H_0$  which, on the classical level, defines an integrable system; the classical phase space then is completely filled out by invariant tori [1]. We now apply a perturbation to our system, i.e., we consider the Hamiltonian

$$H^{\lambda} = H_0 + \lambda H_{\text{int}} , \qquad (1.1)$$

where the coupling strength  $\lambda$  varies in time:  $\lambda = \lambda(t)$ . Now, when  $\lambda$  is slowly increased from zero, the system will, in general, not remain integrable and resonances that are bounded by separatrices appear in the classical phase space. According to the Kol'mogorov-Arnol'd-Moser (KAM) theorem [2], invariant tori still exist and form a set with large measure, at least for small  $\lambda$ , and a trajectory initially confined to an  $H_0$  torus with action  $I_i$ will tend to stay on a torus with almost the same action when  $\lambda$  moves in time. However, this adiabatic invariance of the action breaks down if a separatrix is crossed: Separatrix crossing leads to a "jump" of the adiabatic invariant [3,4]. A classical state of our system (We define a classical state as an ensemble of points in phase space equally distributed on an invariant torus at  $\lambda=0$ .) has been "captured" by a resonance and has changed its topology. In a semiclassical approach to the corresponding quantum system [5], eigenstates are obtained by quantizing invariant classical tori by the Bohr-Sommerfeld rules or its generalizations, the Einstein-Brillouin-Keller (EBK) conditions [6–8]. Thus, a classical transition from an  $H_0$  torus with action  $I_i$  to a state with final action  $I_f$ reached by separatrix crossing "corresponds" to a transition in the quantum system.

An interesting phenomenon occurs if  $\lambda(t)$  has the form of a smooth pulse. The initial torus is then caught and subsequently released by the resonance. During the release the caught manifold passes simultaneously the two branches of the separatrix and thus, finally, escapes to two different  $H_0$  tori, i.e., to two classical states. Quantum mechanically speaking, the pulse has induced a transition to a superposition of two stationary states; their relative weights and the phase relation can be inferred semiclassically. Clearly, this mechanism appears in some sense analogous to transitions induced by "avoided" level degeneracies.

To make these analogies and connections more transparent is the main purpose of the present work. But keeping in mind Arnol'd's widely quoted statement that dynamical systems with even only two degrees of freedom lie beyond our present mathematics, it is obvious that this is not an easy enterprise. Therefore, we do not aim at mathematical rigor but rather try to make the physical content of the above-sketched scenario as clear as possible. To this end, we shall proceed as follows. In the second section, we give a more detailed description of the essential mechanism, together with a numerical example which clearly shows how "classical transitions" occur in a generic system. In Sec. III we discuss recently published experimental measurements from our point of view and show that there is strong evidence for transitions induced by separatrix crossing. The final section contains a critical discussion of the mechanism presented above only in an introductory, simplified, or even oversimplified version.

# II. TRANSITIONS DUE TO SEPARATRIX CROSSING – A NUMERICAL STUDY OF THE GENERIC CASE

To begin with, we consider a classical integrable Hamiltonian system with Hamiltonian function  $H_0$ . The

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phase space  $\{(p,x)\}$  then is completely stratified into tori which are invariant under the flow generated by  $H_0$  [1]. In the case of only one degree of freedom, the invariant tori are topologically equivalent to circles. If we then add a perturbation  $\lambda H_{int}$  to such a system with one degree of freedom, it remains integrable if the perturbation does not depend on time. But since we are interested in the generic, nonintegrable case, we choose a periodically time-dependent perturbation, i.e., we consider a system with "1.5 degrees of freedom."

For a periodically time-dependent quantum system, the Floquet states and the quasienergies adopt the role which stationary states and energies play in time-independent systems. In the same way that stationary states can semiclassically be calculated by torus quantization, Floquet states can be obtained by quantizing time-periodic vortex tubes [9] in the extended phase space  $\{(p,x,t)\}$ . Thus, in order to transfer the cursory discussion of the preceding section to the present case, the invariant tori have to be replaced by vortex tubes which, for periodically driven one-dimensional systems, are easy to visualize in Poincaré surfaces of section.

Let us assume that the perturbation  $\lambda H_{int}(t)$  introduces a resonance bounded by a separatrix in the classical phase space, as indicated schematically in Fig. 1(a). Now we let the coupling strength  $\lambda$  slowly increase in time, that is, we consider a smooth function  $\lambda(t) \equiv \overline{\lambda}(t/T_a)$ with  $\lambda(0)$  equal to zero and  $T_a$  being a "large" interval of time. As initial conditions we choose an ensemble of trajectories which is confined to a torus  $I_i$  at t=0 with the individual trajectories uniformly distributed in the angle variable. We further assume that the initial action  $I_i$  is, after semiclassical quantization according to the Bohr-Sommerfeld rules, associated to an eigenstate of the quantum system.

By the principle of adiabatic invariance, the action variable will remain approximately constant when  $\lambda$  grows, which means that the trajectories remain close to vortex tubes with action  $I_i$ . On the other hand, the area of the resonance grows [2] when  $\lambda$  is increased. Hence, there will be a critical moment  $t = t_c$  when the separatrix touches the tube with the initial action  $I_i$  [Fig. 1(b)].

Whereas the trajectories have simply wound around their tubes for  $t < t_c$ , they now behave differently: If, for  $t \ge t_c$ , the coupling strength were kept constant at  $\lambda_c \equiv \lambda(t_c)$  and if all trajectories were located exactly on the separatrix, they would finally slow down in the vicinity of the hyperbolic periodic orbit h, since the period of rotation diverges on the separatrix. However, for  $t \approx t_c$ the trajectories are actually found not on, but close to, the separatrix. As soon as they have crossed the separatrix and come close to the hyperbolic periodic orbit, they start to follow the flow in the other direction of the unstable manifold and thereby also become associated to what has been a different tube before. In fact, at  $\lambda = \lambda_c$  two formerly different tubes are coupled by h.

When  $\lambda$  is then increased beyond its critical value, the area of the resonance still grows, and the two vortex tubes that have been coupled at  $t = t_c$ , combine to one "new" tube which is "captured" inside the separatrix



FIG. 1. Separatrix crossing in a periodically driven system with one degree of freedom (schematically). (a) A perturbation  $\lambda H_{int}(t)$  introduces a resonance in a surface of section, i.e., a separatrix with a hyperbolic fixed point h and an elliptic fixed point e. When  $\lambda$  is increased, the area of action enclosed by the resonance (shaded) grows, whereas the trajectories stay close to the vortex tube with the initial action  $I_i$  (inner circle). The arrows indicate the direction of the flow on the separatrix. (b) The separatrix touches the tube that has evolved adiabatically from the initial one: trajectories close to the separatrix start to follow the flow in the other direction of the unstable manifold (indicated by the broken arrow) and also become associated to a tube of different action  $I_f$ . (c) As  $\lambda$  is increased further, the tubes that have been coupled in (b) melt into one which is "captured" inside the separatrix. The trajectories are now found close to this tube. (d) When  $\lambda$  is then decreased back to zero, the separatrix crossing occurs a second time. After the pulse, the action values of the trajectories are centered around  $I_i$  and  $I_f$ : a "classical transition" has taken place.

[Fig. 1(c)]. Denoting the action of the tube that has been coupled to the original one by  $I_f$ , the action of the resulting tube after the separatrix crossing obviously is  $I_m = I_f - I_i$ , and the trajectories now are confined to vortex tubes with actions approximately equal to  $I_m$ .

After the coupling strength has reached its maximal value, we let  $\lambda$  slowly decrease again. At  $\lambda = \lambda_c$ , the separatrix crossing occurs a second time, and the trajectories are distributed from  $I_m$  to the two tubes with actions  $I_i$  and  $I_f$ . This situation is then adiabatically transported to  $\lambda = 0$  [see Fig. 1(d)].

Now, according to our assumptions, the initially "populated" torus  $I_i$  is semiclassically associated to a quantum eigenstate of  $H_0$ , and we stipulate also that the final torus  $I_f$  correspond to an eigenstate. In this case, the correspondence principle strongly suggests that the classical transition from  $I_i$  to  $I_f$  should have its counterpart in quantum mechanics.

However, in the discussion above we have ignored the effect of stochastic motion: In a near-integrable system, a resonance is not simply bounded by a separatrix but rather by a stochastic layer originating from homoclinic tangles close to hyperbolic periodic orbits. Nevertheless, the following numerical example will show that, despite all complications, our schematic discussion remains valid also in the case of partly stochastic motion. We take a system with "1.5 degrees of freedom," namely, a periodically driven one-dimensional Morse oscillator, the Hamiltonian of which can be written as

$$H^{\Lambda}(\tau) = \frac{P^2}{2m} + D[1 - \exp(-\beta X)]^2 + \Lambda X \cos\Omega\tau . \qquad (2.1)$$

Scaling the variables according to

$$x = \beta X ,$$
  

$$p = P / \sqrt{2mD} , \qquad (2.2)$$

 $\omega t = \Omega \tau$ 

with

$$\omega \equiv \Omega \left[ \frac{m}{2D\beta^2} \right]^{1/2}, \qquad (2.3)$$

the Hamiltonian can be expressed in dimensionless quantities to read

$$H^{\lambda}(t) = \frac{1}{2}p^{2} + \frac{1}{2}(1 - e^{-x})^{2} + \lambda x \cos \omega t$$
 (2.4)

with a scaled coupling strength  $\lambda$  given by

$$\lambda = \frac{\Lambda}{2D\beta} . \tag{2.5}$$

Note that the classical action scales with a factor

$$\alpha = \left[\frac{2mD}{\beta^2}\right]^{1/2} \tag{2.6}$$

under the transformation (2.2). For instance, for a Morse oscillator with parameters describing an O—H bond [10] one finds

$$\alpha = 22.082\hbar$$
 (2.7)

This number will be relevant when we discuss the correspondence of classical and quantum mechanics.

As is well known, for  $\lambda=0$  the Bohr-Sommerfeld quantization yields the exact quantum-mechanical energy eigenvalues of the unperturbed Morse oscillator

$$H_0 = \frac{1}{2}p^2 + \frac{1}{2}(1 - e^{-x})^2 .$$
 (2.8)

For a fixed nonvanishing value of  $\lambda$ , the Hamiltonian (2.4) depends periodically on time and the Floquet states can be calculated semiclassically by vortex-tube quantization [9]. The association of quantum-mechanical Floquet states of a driven Morse oscillator to classical vortex tubes has been illustrated numerically in Ref. [11].

Figure 2 shows a surface of section of the system (2.4)taken at integer multiples of  $T = 2\pi/\omega$  for  $\lambda = 0.02712$ and  $\omega = 0.8868$  which has been plotted in action-angle variables  $(I, \theta)$  of the unperturbed oscillator (2.8). The dominating structure seen in this figure is the elliptic island that has originated from the principal (1:1) resonance, together with a surrounding stochastic layer. This is a realization of the abstract situation discussed before. If the ratio of the external frequency  $\omega$  and the internal frequency  $\partial H_0 / \partial I$  is 1:1 and if  $\lambda$  is small enough, then the Poincaré-Birkhoff theorem guarantees the existence of one stable (elliptic) and one unstable (hyperbolic) periodic orbit, which appear as elliptic and hyperbolic fixed points in a surface of section [cf. the schematic drawing in Fig. 1(a)]. When  $\lambda$  is increased, the area of the regular island around the elliptic fixed point grows; on the other hand, apparently stochastic motion due to homoclinic tangles at the hyperbolic fixed points also becomes more pronounced.

Let us now turn to the case of a continuously varying coupling strength  $\lambda(t)$  which we specify as

$$\lambda(t) = \lambda_{\max} \sin^2 \left[ \frac{\pi t}{T_a} \right], \quad 0 \le t \le T_a \quad . \tag{2.9}$$

We then fix a set of 1000 initial values which are characterized by a common action  $I_0 = 0.02264$  and uniformly distributed in the angle variable  $\theta$  at t = 0. In order to investigate the time evolution of this ensemble under the influence of a pulse (2.9), we integrate the equations of



FIG. 2. Poincaré surface of section for the driven Morse oscillator (2.4), taken at integer multiples of  $T=2\pi/\omega$ . The parameters are  $\lambda=0.02712$  and  $\omega=0.8868$ .



FIG. 3. Time evolution of an ensemble of trajectories with common initial action  $I_0=0.022$  64 under the influence of a pulse (2.9) with length  $T_a=105T$ . (a) Ensemble at t=30T. (b) t=40T. (c) t=50T. The separatrix crossing has taken place. (d) t=60T. (e) t=70T. The second separatrix crossing occurs. (f) t=80T. (g) Ensemble after the pulse at  $t=T_a$ .

motion numerically. We choose the parameters  $T_a = 105T$ ,  $\omega = 0.8868$ , and  $\lambda_{max} = 0.02727$  so that Fig. 2 visualizes the structure of the phase space at t = 50T close to the moment of maximal coupling strength.

The reason for the particular choice of  $I_0$  and  $\omega$  is that for a Morse oscillator modeling an O—H bond [see (2.7)] the initial action  $I_0$  semiclassically yields the quantummechanical ground state and the frequency  $\omega$  allows for a "four-photon-transition" from the ground state with energy  $E_0$  to the fourth excited state with energy  $E_4$ , i.e., we have  $E_4 - E_0 = 4\hbar\omega$ .

Figure 3(a) shows the ensemble after 30 periods, a situation which roughly corresponds to Fig. 1(a). Up to now, the time evolution has been almost adiabatic. All members of the ensemble approximately lie on a curve which represents a vortex tube of the instantaneous system, i.e., of a Morse oscillator drive with *constant* amplitude  $\lambda_{30T} \equiv \lambda(30T)$ . Conservation of action means that this (possibly only approximately defined) vortex tube is still characterized by an action equal to  $I_0$  with respect to the (approximate) action-angle variables at  $\lambda = \lambda_{30T}$ . But as Fig. 3(a), as well as the following figures, is plotted in action-angle variables defined at  $\lambda = 0$ , conservation of action is not immediately visible.

Ten periods later [see Fig. 3(b)] the effect of the principal resonance begins to show. The bending of the ensemble that can be observed at the right margin indicates the influence of the hyperbolic fixed point located at I=0.084,  $\theta=\pi$ . This situation matches Fig. 1(b) of our schematic discussion.

Again ten periods later [Fig. 3(c)], when  $\lambda \approx \lambda_{max}$ , the separatrix crossing has taken place; the ensemble approximately lies on a vortex tube "inside" the resonance, as in Fig. 1(c).

After 60 periods [Fig. 3(d)], the coupling strength  $\lambda$  is decreasing again. The ensemble still encircles the resonance, showing a pronounced influence of homoclinic points.

The second separatrix crossing occurs at  $t \approx 70T$  [Fig. 3(e)], and ten periods later [Fig. 3(f)] the ensemble apparently has divided into two parts.

Finally, at the end of the pulse [Fig. 3(g)] we find the actions of the individual trajectories centered around two values: the action  $I_0$  of the initial state and the action  $I_f = 0.2031$ .

At this stage of discussion, an important remark is appropriate. The manifold on which all initial values of our model calculation lie is a circle in the phase space and, strictly speaking, this manifold remains a circle under the Hamiltonian flow. But the initial circle is stretched and folded so strongly [see, in particular, Fig. 3(c)] that it is almost impossible to recognize the fact that after the pulse all ensemble points still lie on a circle in the topological sense; it appears more effective to describe the resulting ensemble in terms of two disjoint sets. In this sense, a "classical transition" has taken place. This point of view is clearly vindicated by Fig. 4 which shows a histogram of the final actions of all trajectories.

It is of interest now to note that  $I_f$  is almost exactly equal to the action  $I_4 = 0.2038$  which semiclassically cor-



FIG. 4. Histogram of the final action values of the trajectories after the pulse [see Fig. 3(g)]. The arrows indicate the actions which semiclassically yield the bound states of a quantum-mechanical Morse oscillator modeling an O—H bond.

responds to the fourth excited state of an O—H Morse oscillator. Hence, one is led to expect that for this particular example the classical transition corresponds to a transition from the ground state to the fourth excited state in the quantum system.

Indeed, this expectation is confirmed by quantummechanical calculations which model the dynamics of molecular vibrations interacting with a resonant, short laser pulse [10,12]. The interpretation of the transitions found in these studies has been formulated in terms of "splitting" of a wave function, adiabatic evolution, and interference of Floquet states [12,13]. If the laser frequency is chosen such that the initial and final states are "resonant," their instantaneous quasienergies are degenerate at the initial stage of the pulse. Hence, the wave function of the initial state splits into a superposition of two Floquet states; both parts of the wave function then evolve adiabatically until they interfere at the end of the pulse. Obviously, the violation of adiabaticity caused by the degeneracy of quasienergies is matched in classical mechanics by the intrinsic nonadiabaticity at the moment of separatrix crossing.

To finish the discussion of the model calculations, we illustrate the role of the pulse length  $T_a$  and show in Fig. 5 both the fraction  $p^+$  of trajectories which have a final action close to  $I_4$  and the rms deviation from the mean final action of that fraction as functions of  $T_a$ . The oscillations of  $p^+$  are caused by the fact that the individual trajectories circle around the elliptic periodic orbit as long as they are "inside" the resonance. The increasing sharpness of the final distribution observed for  $T_a < 100T$ indicates increasingly adiabatic behavior, the fact that the rms deviation does not decrease further for  $T_a > 100T$ but rather becomes approximately stationary stems from the stochastic part of the dynamics. In essence, the basic transition mechanism determined solely by the 1:1 resonance is valid in the whole range  $100T \le T_a \le 300T$  plotted in Fig. 5.

Let us finally discuss the question under which conditions classical transitions involve actions  $I_1$  and  $I_2$  which



FIG. 5. Fraction  $p^+$  of trajectories with a final action close to  $I_4$  (solid line) and rms deviation  $\sigma$  from the mean action of that fraction (dotted line) as functions of the pulse length  $T_a$ .

actually both correspond to quantum-mechanical eigenstates  $|n_1\rangle$  and  $|n_2\rangle$  (we assume  $n_2 > n_1$ ) of a system described by a Hamiltonian *H*. Let  $I_k = \hbar(n_k + \frac{1}{2})$  and  $E_k = H(I_k)$  for k = 1, 2. Then, for  $m \equiv n_2 - n_1$ , choose the frequency  $\omega$  such that  $E_2 - E_1 = m \hbar \omega$  and define  $I_{\text{res}}$ by

$$\frac{\partial H(I_{\rm res})}{\partial I} = \omega . \qquad (2.10)$$

The assumption that H(I) can be quadratically approximated around  $I = I_{res}$  (in the case of the Morse oscillator H actually is a quadratic function of I) then yields

$$I_{\rm res} = \frac{1}{2}(I_1 + I_2) \tag{2.11}$$

or, equivalently,

$$(I_2 - I_{\rm res}) = (I_{\rm res} - I_1)$$
 (2.12)

If we further assume that the area of the principal 1:1 resonance located at  $I=I_{res}$  grows symmetrically to higher and lower actions I when the coupling strength  $\lambda$  is increased (which is a reasonable assumption as long as  $\lambda$ does not become excessively high), trajectories with initial action  $I_1$  or  $I_2$  will enter the resonance at the same value of  $\lambda$ . This fact implies that the final action values of an ensemble with common initial action  $I_i = I_1$  are centered around  $I_1$  and  $I_2$ . A classical transition from  $I_1$ to  $I_2$  takes place.

These considerations are also interesting from a different point of view: An important question in the study of atoms or molecules interacting with short laser pulses is how to choose the laser frequency in order to achieve a certain transition with the highest possible efficiency. Now, classically it is the principal 1:1 resonance which favors a transition by separatrix crossing most strongly, in the sense that is covers the largest area in phase space at a given value of the coupling strength. Hence, assuming that the classical-quantum correspondence holds, a transition from a quantum state  $|n_1\rangle$  to  $|n_2\rangle$  is favored most if the frequency is chosen such that both states are classically coupled by the principal reso-

nance. Contrary to what might be expected by perturbative arguments, this line of reasoning implies that for transitions induced by pulses it is not the "one-photon transition" which is the most effective, but rather the optimal frequency satisfies

$$E_2 - E_1 = (n_2 - n_1)\hbar\omega$$
; (2.13)

i.e., the difference in quantum number should be equal to the "number of absorbed photons." It is interesting to note that indications for this phenomenon have been seen in Ref. [10].

## III. EVIDENCES FROM EXPERIMENTAL DATA — MICROWAVE EXPERIMENTS REEXAMINED

Highly excited hydrogen atoms exposed to pulsed microwave radiation [14,15] are quite attractive systems for an experimental study of the perturbative and nonperturbative dynamics of periodically driven quantum systems, mainly because all relevant parameters can be accurately controlled. In particular, there are recent measurements performed with an experimental setup where highly excited hydrogen atoms were exposed to both a pulse of microwaves and an additional static electric field, with the microwave field being linearly polarized in the static-field direction [14]. The strength  $F_s$  of the static field was chosen to be even higher than that of the microwave field, so that the atoms are strongly elongated in one direction and behave essentially as driven onedimensional systems that can be described by a Hamiltonian

$$H^{\lambda(t)}(t) = H_0 + \lambda(t)x \sin\omega t , \qquad (3.1)$$

where  $\omega$  denotes the microwave frequency and

$$H_0 = \frac{p^2}{2} + V(x) - F_s x \tag{3.2}$$

with

. ...

$$V(x) = \begin{cases} -\frac{1}{x}, & x > 0 \\ \infty, & x \le 0. \end{cases}$$
(3.3)

It is interesting to note that in this case also the classical internal frequency

$$\Omega = \frac{\partial H_0}{\partial I} \tag{3.4}$$

can be adjusted by suitably tuning the static field  $F_s$ . To first order in  $F_s$ , one has

$$\Omega = \frac{1}{n^3} - 3nF_s , \qquad (3.5)$$

where the classical action I has been replaced by the quantum number n. We remark that a proper treatment of the Stark effect in hydrogen [16] leads to the same expression for high quantum numbers, if one considers only those parabolic substates which are polarized most strongly in the direction of the static field.

The shape function  $\lambda(t)$  in (3.1) and, hence, the time

scale  $T_a$  is fixed by the fact that in the experiments under consideration [14] a fast beam of excited atoms transverses a rectangular waveguide operated in the  $TE_{10}$ mode and the atoms experience essentially a half-sinewave envelope

$$\lambda(t) = \lambda_{\max} \sin\left[\frac{\pi t}{T_a}\right], \quad 0 \le t \le T_a \tag{3.6}$$

which is slightly modified near t=0 and  $t=T_a$  by the influence of the entrance and exit holes on the field distribution in the waveguide. This modification may be of importance because the smoothness of the actual shape function, that is, the number of vanishing derivatives at the end points, determines both in classical and quantum mechanics the magnitude of nonadiabatic effects [17].

From Eq. (3.5) it follows immediately that the "scaled frequency," i.e., the winding number  $\gamma$ , is given by

$$\gamma = \frac{\omega}{\Omega} = \frac{n^3 \omega}{1 - 3n^4 F_s} . \tag{3.7}$$

Therefore,  $\gamma$  is larger than  $n^3\omega$ , which is the scaled frequency in the case of a vanishing static field.

In actual experiments, the shift of the scaled frequency due to the static field can be quite strong. For instance, in the situation of Fig. 1(c) of Ref. [14] the measurements were carried out at  $\omega/2\pi = 13.00$  GHz with  $F_s = 6.25$ V/cm and a microwave amplitude  $\lambda_{max}$  of merely 3.44 V/cm; the interaction time  $T_a$  was roughly 100 microwave cycles. For this particular example and  $n_i = 72$ , we have (in atomic units)  $n_i^3 \omega = 0.737$  [14] compared to  $\gamma = 0.818$ . In addition, one finds

γ	
0.899	
0.942	
0.987	
1.035	
	γ 0.899 0.942 0.987 1.035

Whereas the state closest to the classical principal 1:1 resonance  $(\gamma = 1)$  is  $n_{\rm res} = 80$  if no static field is present, we now have  $n_{\rm res} = 76$ . The experimental data displayed in Fig. 1(c) of Ref. [14] show a pronounced double peak, one maximum of excitation probability after the interaction being found at the initial state  $n_i = 72 = (n_{\rm res} - 4)$ , the other maximum at  $n_f = 80 = (n_{res} + 4)$ . Thus, the experimental result shares the main feature of our model calculations discussed in Sec. II. After a state initially close to a prominent classical resonance has interacted with an external periodic field whose amplitude changes continuously in time, the final population of states is a distribution with two maxima which lie symmetrically around the position of the resonance. It appears safe to conclude that the measurements summarized in Fig. 1(c) of Ref. [14] reflect the effect of classical separatrix crossing on a quantum system, that is, the effect of prominent classical resonances capturing and releasing states. For the time scales involved this mechanism is dominated by regular motion and stochasticity generated by homoclinic points is only of secondary importance. Nonetheless, we do not

exclude a possible role of localization phenomena [18] in building up a nonresonant background.

In a similar manner, one can also analyze the other measurements published in Ref. [14]. To obtain the data of Fig. 1(a), the initial state again was  $n_i = 72$  and the same static field  $F_s = 6.25$  V/cm was used together with a microwave field of frequency  $\omega/2\pi = 18.00$  GHz. The initial state closest to the main resonance then is  $n_{\rm res} = 69$ :

n	γ	
68	0.933	
69	0.980	
70	1.028	
71	1.079	
72	1.132	

In this case, the initial state  $n_i = 72$  is located "above" the center of the classical resonance and, hence, one has to expect a deexitation to lower states. This is precisely what has been observed: the experimental data show a maximum at n = 66. Note that (72-69)=(69-66).

Finally, under the conditions of Fig. 1(b) of Ref. [14] the initial state itself is close to  $\gamma = 1$ . In such a case, neither excitation nor deexcitation is favored classically and, indeed, experimentally one finds only one maximum of the final population probability at exactly  $n_i = 72$ .

From a purely quantum-mechanical point of view [19], it is the structure of the quasienergy spectrum that determines which transitions are favored. For highly excited hydrogen atoms, the effect of the mod $\omega$  operation which maps the unperturbed energy eigenvalues at  $\lambda=0$  into the Brillouin zone is easy to oversee. The classicalquantum correspondence is manifested by the fact that, for small *m*, the eigenvalues of states  $(n_{\rm res} - m)$  and  $(n_{\rm res} + m)$  are mapped close to each other such that Landau-Zener transitions among these states are possible when the microwave amplitude varies. For a numerical example of such a situation, see Figs. 6 and 7 of Ref. [19]. In the situation discussed there, one has  $n_{\rm res} = 75$ ,  $n_i = 72$ and favored transitions to  $n_f = 78$  and  $n_f = 79$ , so that the characteristic relation  $(n_f - n_{\rm res}) \approx (n_{\rm res} - n_i)$  again holds.

To summarize, the presence of the static field does not alter the basic mechanism, but it allows one to shift the location of the resonance  $\gamma = 1$  in the classical phase space such that it lies close to various invariant manifolds which, after semiclassical quantization, correspond to different initial states; prominent experimentally discovered regularities find a natural explanation by merely observing the classical resonance being located "below," "above," or "close to" the initial state. This simple fact may help to interpret the results of future experiments aimed at a deeper understanding of the classical-quantum correspondence for time-dependent phenomena.

#### **IV. DISCUSSION**

In the preceding sections we developed a simple picture of resonant trapping of classical states and its semiclassical analog in quantum systems; we illustrated its relevance by a numerical analysis of a simple periodically driven system and, finally, pointed out its experimental verification.

There are, however, a few serious objections that may cast considerable doubt on the general validity of this simple picture. We shall try in the following to specify more clearly the conditions under which, nonetheless, a semiclassical translation of separatrix crossing of classical states (i.e., of ensembles of phase space points distributed on initial manifolds) explains prominent features of quantum-mechanical processes.

In particular, the following remarks seem to be of importance.

(i) A system with more than only one degree of freedom will, in general, not remain integrable when the perturbation  $\lambda H_{int}$  is switched on. The set of preserved invariant tori does then not fill out the complete phase space, but rather forms a Cantor set. The question to which extent the principle of adiabatic invariance can be applied to such a situation is a difficult, and yet unsolved, problem (see, however, the recent work by Dana and Reinhardt [17]). In addition, the semiclassical quantization by Bohr-Sommerfeld rules or its generalizations, the EBK conditions, is obviously questionable because precisely the required tori may not exist.

(ii) If, during the variation of a control parameter  $\lambda$ , the action variables of a system with *n* degrees of freedom remain constant, the winding numbers, i.e., the ratios of the relevant *n* frequencies, change continuously. Whenever a winding number hits a rational number, a resonance appears in the classical phase space. Therefore, when  $\lambda$  is varied by a finite amount, the system meets an infinity of resonances.

(iii) In the generic case, a resonance in phase space is bounded not simply by a separatrix but rather by a stochastic layer with a certain extension. This stochastic layer blurs the classical transition in the sense that it leads to a less sharp distribution of final actions of the individual trajectories.

The basic transition mechanism proposed in this paper is expected to survive in the generic case of mixed regular and stochastic dynamics, if stochastic motion predominates only in a small fraction of the relevant part of phase space. That is, the still existing tori should occupy a major part of phase space so that the application of EBK-type quantization procedures makes sense at least approximately and, furthermore, the stochastic layers surrounding the resonances should not become too broad. In this context the following observation provides an interesting hint concerning the assumed regularity of phase space for small and large coupling strength  $\lambda$  [cf. (1.1)]. Using properly scaled variables we have shown that, assuming Hamiltonian  $H_0$  describing anharmonic oscillators, the classical phase space shows a remarkable simple structure for strong driving forces. Stochastic motion is confined to a well-defined full torus (doughnut) surrounded by stable vortex tubes [20]. For small  $\lambda$ , the familiar KAM regularity prevails. Thus, both regimes are accessible for EBK quantization.

It appears that a more severe difficulty is connected with (ii). Whereas we have assumed that it suffices to take into account the effect of basically one isolated hyperbolic fixed point, it should be clear that each of the infinitely many resonances which the system encounters during the parameter variation introduces its own hyperbolic and elliptic fixed points, as shown by Poincaré and Birkhoff.

It is at this point that we have to observe the decisive role played by the relative magnitude of time scales involved: pulse length and inverse frequency of the driving force versus internal time scales of the system. More specifically, we argue as follow.

When specifying the exact way in which the parameter  $\lambda$  varies in time, we necessarily have to introduce a certain characteristic time scale  $T_a$ . On the other hand, each resonance is characterized by its own intrinsic time scale which may be defined, for instance, by the time it takes to stretch the flow in the direction of the unstable manifold by a given factor. The condition for a certain resonance to be "effective," that is, the condition that it actually induces a classical transition, obviously is that its own intrinsic time scale be comparable with the interval of time this very resonance is "seen" by the trajectories when  $\lambda$  varies; this interval, in turn, is determined by  $T_a$ . In their study of adiabatic invariance in the standard map, Dana and Reinhard [17] have estimated the typical time scale associated with a resonance caused by a rational winding number p/q and have found it to be an increasing function of the denominator q. It is highly plausible to assume that this result is valid also in the general case. Hence, once the time scale  $T_a$  is fixed, only a finite number of potentially effective resonances remain, and, under favorable conditions, it is possible that essentially only one of them determines the final action distribution [17]. It should be noted that this discussion of a purely classical phenomenon parallels our introduction of a coarse-graining procedure for the quasienergy spectrum of a quantum system [21].

Certainly, there are many more questions, most of which cannot be answered at the present time. Nevertheless, we hope to have illustrated the role of (primary) classical resonances for a description of transitions in the corresponding quantum system which are induced by the time dependence of external parameters or, rather, by short-pulsed interactions.

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