Brief Reports

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Resonance overlap structure in the microwave ionization of the hydrogen atom

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The microwave ionization of the hydrogen atom involves most of the open issues concerning classical and quantum chaos. Much recent research has considered quasi-one-dimensional extended states for which ionization thresholds have been estimated using a classical picture in which ionization proceeds through the overlap of an infinity of nonlinear resonances. Using a canonical transformation to Deprit's "Lissajous elements" which makes the two-dimensional nature of the problem explicit, an accurate and *improved* ionization threshold, compared to previous resonance overlap criteria, is obtained through the overlap of only two nonlinear resonances in the one-dimensional limit.

The microwave ionization of the hydrogen atom (MIH) is currently of considerable experimental and theoretical interest from a number of standpoints.[1-3] Apart from the insight the problem might provide into the general behavior of Rydberg atoms in external fields, the system allows a unique opportunity to compare the predictions of classical and quantum mechanics with experiment. Evidence is mounting rapidly that quantum mechanics at least suppresses the chaos which is dominant in classical mechanics, and may, in fact, eliminate chaos altogether. Support for this is provided by recent calculations by Casati *et al.*[4,5] on the MIH which suggest the existence of a frequency window in which the "classical" electron ionizes while the quantum electron does not. This is surprising since for the rather high quantum numbers involved the electron is expected to behave classically. Consequently there is a pressing need to perform the corresponding experiment, and considerable effort is being directed to this end.[2]

Most of the theoretical studies of the MIH problem have considered one-dimensional models in order to simplify the calculations. This is a reasonable assumption because many of the experiments themselves consider extended, quasi-one-dimensional, hydrogen atoms. Classically, however, the overlap of resonances in the *three*-dimensional MIH system is expected to be more important and to intensify the effect of chaos on the ionization thresholds. It is therefore important to develop theoretical models, both classical and quantal, to deal with the fully three-dimensional MIH system. Indeed, recent experiments have begun to study regimes where a three-dimensional theory is necessary.[2,6] Yet, even in the one-dimensional case the correspondence between the classical and quantum dynamics is far from understood. The purpose of this communication is to present a new approach to the prediction of the one-dimensional classical ionization threshold which is in better agreement with the exact dynamics than previous estimates,[7] provides a much simpler intuitive picture of the onset of chaos, and, importantly, is expected to be readily extendible to treating the three-dimensional problem.

The paper is organized as follows: after introducing the classical Hamiltonian, a change of coordinates is made to reduce the system to two dimensions, after which a canonical transformation to extended phase space, followed by a further transformation to action-angle variables results in a Hamiltonian whose resonance structure is particularly apparent. Specializing to a one-dimensional model corresponding physically to highly extended states, an explicit prediction of the classical ionization threshold is made. The prediction is compared with and found to be in excellent agreement with the exact dynamics. Finally, ramifications of the study for the three-dimensional problem are briefly discussed.

The classical Hamiltonian in atomic units is

$$H=\tfrac{1}{2}(P_x^2+P_y^2+P_z^2)-\frac{1}{r}+\varepsilon z\cos(\omega t)\ , \tag{1}$$

where ε is the field strength and ω the field frequency. Due to a well-known scaling property of the Kepler problem, the classical dynamics depends only on the quantities[3]

$$\varepsilon_0=\varepsilon n_0^4, \omega_0=\omega n_0^3 , \tag{2}$$

where n_0 is the principal quantum number of the initial state. This considerably simplifies the classical studies since the dynamics is now independent of n_0 and only ε_0 and ω_0 need be varied.

Because the problem has axial symmetry, the magnetic quantum number m is preserved and the Hamiltonian may be reduced to two dimensions by a transformation to squared parabolic coordinates which are commonly employed in studying the dc Stark effect,[8]

$$x=uv\cos\varphi, \quad y=uv\sin\varphi, \quad z=\tfrac{1}{2}(u^2-v^2) , \tag{3}$$

giving

$$H=\frac{1}{2(u^2+v^2)}\left[P_u^2+P_v^2+\frac{m^2(u^2+v^2)}{u^2v^2}-4+\varepsilon(u^4-v^4)\cos(\omega t)\right] , \tag{4}$$

where the parabolic quantum numbers n_u, n_v, and m are related to the principal quantum number n by the relation[8]

$$n=n_u+n_v+|m|+1 . \tag{5}$$

Just like the original Hamiltonian (1), the Hamiltonian (4) has a singularity at the origin which leads to numerical instabilities in the integration of Hamilton's equations of motion. The unphysical singularity may be removed by using the technique of regularization developed in celestial mechanics.[9] This is implemented by first eliminating the time dependence in Eq. (4) by a transformation to extended phase space, followed by an implicit change of the time variable (details are given by Szebehely[9]). The excursion to extended phase space is effected by the following canonical transformation:

$$P_\theta=-E, \quad \theta=t , \tag{6}$$

where E is the energy. Multiplication by (u^2+v^2) (equivalent to a time transformation) completes the regularization, giving

$$\bar{H}=2=K_1+K_2+\frac{\varepsilon}{2}(u^4-v^4)\cos(\omega\theta) , \tag{7}$$

where

$$K_1=\frac{1}{2}\left[P_u^2+\frac{m^2}{u^2}+2b^2u^2\right] , \tag{8a}$$

$$K_2=\frac{1}{2}\left[P_v^2+\frac{m^2}{v^2}+2b^2v^2\right] , \tag{8b}$$

and

$$2b^2=P_\theta=-E . \tag{9}$$

Apart from the centrifugal terms the Hamiltonian resembles that of two nonlinearly coupled oscillators. Since much of classical perturbation theory and the theory of resonance overlap has been developed in the context of coupled oscillators it would be particularly convenient to develop a transformation to action-angle variables which exploits the intimate connection between the hydrogen atom and the isotropic oscillator. Deprit[10] has recently obtained such a transformation which deals quite elegantly with the centrifugal term. The canonical transformation to Deprit's "Lissajous" elements, i.e., $(u,P_u)\rightarrow(\phi,\Phi)$, and $(v,P_v)\rightarrow(\psi,\Psi)$ is such that

$$K_1\rightarrow 2b\Phi , \tag{10a}$$

$$K_2\rightarrow 2b\Psi . \tag{10b}$$

The generator of the transformation W is of the F_2 type (i.e., a function of old coordinates and new momenta) and satisfies

$$P_u=\frac{\partial W}{\partial u}, \quad \phi=\frac{\partial W}{\partial\Phi} , \tag{11}$$

and similarly for the $(v,P_v)\rightarrow(\psi,\Psi)$ transformation. (The Hamiltonian is symmetric in u and v so it is necessary to describe the transformation for only one set of variables.) The required generating function $W(u,\Phi)$ is obtained from the Hamilton-Jacobi equation

$$\frac{1}{2}\left[\frac{\partial W}{\partial u}\right]^2+\frac{m^2}{2u^2}+2b^2u^2=2b^2\Phi , \tag{12}$$

which, solving for $W(u,\Phi)$, gives

$$W(u,\Phi)=\int_{u_1}^{u}du\left[4b\Phi-4b^2u^2-\frac{m^2}{u^2}\right]^{1/2} , \tag{13}$$

where the lower limit is the positive root of the integrand. Defining the quantity

$$f(\Phi,m)=\left[1-\left[\frac{m}{\Phi}\right]^2\right]^{1/2} \tag{14}$$

and using Eq. (9) it can be shown straightforwardly that

$$u^2=\frac{\Phi}{2b}[1-f\cos(2\phi)], \quad uP_u=\Phi f\sin(2\phi) , \tag{15}$$

which changes K_1 into $2b\Phi$. Similarly, defining a quantity $g(\Psi,m)$ in analogy with $f(\Phi,m)$ in Eq. (14), followed by the transformation, changes K_2 into $2b\Psi$. After these transformations, the Hamiltonian in Eq. (7) becomes

$$\bar{H}=2=2b(\Phi+\Psi)+\frac{\varepsilon}{2}\cos(\omega\theta)\left[\left[\frac{\Phi}{2b}\right]^2[1-f\cos(2\phi)]^2-\left[\frac{\Psi}{2b}\right]^2[1-g\cos(2\psi)]^2\right] . \tag{16}$$

In the absence of a field this Hamiltonian resembles that for two degenerate uncoupled harmonic oscillators in

action-angle variables with frequency $4b$, i.e.,

$$2=2b(\Phi+\Psi)\ , \tag{17}$$

and thus

$$b=\frac{1}{\Phi+\Psi}\ , \tag{18}$$

which in the light of Eq. (9) leads directly to the quantization condition

$$\Phi+\Psi=2n,\quad n=1,2,3,\ldots\ . \tag{19}$$

At this point an approximation is introduced; Eq. (18) is substituted into the Hamiltonian in Eq. (16) for all terms in b occurring *in the perturbation,* after which the resulting equation is rearranged, giving

$$\beta\approx\frac{-2}{(\Phi+\Psi)^2}+\varepsilon\left[\frac{\Phi}{2}\right]^2[1-f\cos(2\phi)]^2\cos(\omega\theta)+\varepsilon\left[\frac{\Psi}{2}\right]^2[1-g\cos(2\Psi)]^2\cos(\omega\theta)\ , \tag{20}$$

where β is the total energy.

The leading order term in b is given by Eq. (18) and this approximation is expected to be reasonable even in the presence of the perturbation. The structure of the new Hamiltonian β is particularly appealing since it shows clearly how the two oscillators are coupled to the perturbation, as well as indirectly to each other by the perturbation. To consider extended, quasi-one-dimensional states, terms in Ψ and ψ are omitted, giving

$$\beta=\frac{-2}{\Phi^2}+\frac{\varepsilon}{4}\Phi^2\left[1+\frac{f^2}{2}-2f\cos(2\phi)+\frac{f^2}{2}\cos(4\phi)\right]\times\cos(\omega\theta)\ . \tag{21}$$

After a further canonical transformation,

$$(2\phi,\Phi)\rightarrow(\lambda,2n)\ , \tag{22}$$

the Hamiltonian becomes

$$\beta=\frac{-1}{2n^2}+\varepsilon n^2\cos(\omega\theta)\left[1+\frac{f^2}{2}-f\cos\lambda+\frac{f^2}{2}\cos(2\lambda)\right]\ . \tag{23}$$

This Hamiltonian is quite analogous to that studied by Delone *et al.*[11] except that the resonance overlap structure of the problem may be analyzed directly using the Hamiltonian as it stands without the need to perform a Fourier expansion of the perturbation and consider, in principle, the overlap of an infinity of classical resonances. The perturbation in Eq. (23) in fact resembles the first two terms in a Fourier expansion, which expansion would have been necessary had the approximation used to obtain Eq. (20) not been made. Note that the Hamiltonian contains only *two* resonance terms as opposed to the infinite number obtained by Fourier expansion. Whenever the frequency of the external field ω is of the order of the natural Kepler frequency Ω, the electron will be driven resonantly. The kth Kepler frequency of the *unperturbed* electron in terms of the corresponding principal quantum number n_k is given by

$$\Omega_k\sim n_k^{-3}\ , \tag{24}$$

and so the resonance condition becomes

$$\omega=k\Omega_k=kn_k^{-3}\ . \tag{25}$$

The critical field for chaos corresponds to the situation where the resonances start to overlap. Expanding the first term in the Hamiltonian around the resonance center in the standard way[11,12] leads to a pendulumlike Hamiltonian, from which the widths of the two resonances can easily be calculated,

$$\Delta\nu_1=\frac{2}{n_1}\left[3\varepsilon\frac{f}{2}\right]^{1/2},\quad \Delta\nu_2=\frac{2}{n_2}(3\varepsilon f^2/8)^{1/2}\ , \tag{26}$$

and the overlap criterion may be formulated in terms of the ratio of the island width to the separation,[12] i.e.,

$$s=\frac{\Delta\nu_1+\Delta\nu_2}{\Omega_1-\Omega_2}\ . \tag{27}$$

If $s>1$ the resonances start to overlap and, according to empirical but generally applicable criteria obtained by

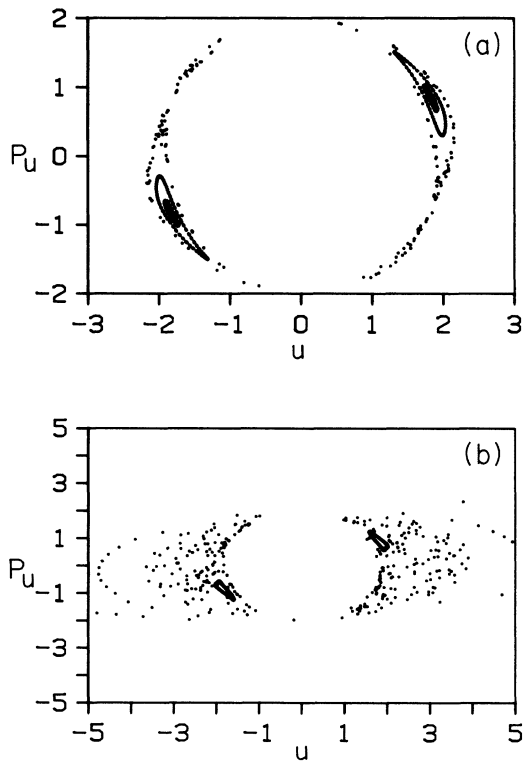

FIG. 1. Poincaré surfaces of section corresponding to $\omega_0=1$ and the field strengths (a) $\varepsilon_0=0.021$ and (b) $\varepsilon_0=0.053$. In each case six *representative* trajectories with different initial conditions have been integrated. In (a) phase space is largely filled with tori; however, close to the separatrix there is some chaos developing, indicating that the resonances are starting to overlap. In (b) most of phase space is chaotic and many of the classical trajectories eventually ionize.

Chirikov,[12] when $s \approx 2.5$ the resonances are strongly overlapping and widespread chaos is apparent. Applying Chirikov's overlap criteria leads to the critical fields

$$\varepsilon_{\text{crit}}^{s=1} \approx \frac{1}{47n^4}, \quad \varepsilon_{\text{crit}}^{s=2.5} \approx \frac{1}{19n^4}, \tag{28}$$

or, alternatively, $\varepsilon_0^{s=1} \approx 0.021$ and $\varepsilon_0^{s=2.5} \approx 0.053$.

Figure 1 displays Poincaré surfaces of section for the two critical-field strengths (corresponding to $s=1$ and 2.5). Excellent and improved[7] agreement (compared to previous resonance-overlap estimates) between the resonance overlap prediction and the exact dynamics is obtained.[3,11,13] However, a more significant aspect of the method is the simple picture painted in terms of overlap of only two classical resonances. For the full three-dimensional problem, there are two resonances in each of the degrees of freedom, and the form of the Hamiltonian in Eq. (20) lends itself to application of a renormalization approach like that developed by Escande and Doveil.[4] Work is in progress to apply this technique in order to predict the ionization threshold for the fully three-dimensional system.

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