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# Threshold ionization dynamics of the hydrogen atom in crossed electric and magnetic fields

T. Uzer
*School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332-0430*

David Farrelly
*Department of Chemistry, Princeton University, Princeton, New Jersey 08544-1009*
*and Department of Chemistry and Biochemistry, Utah State University, Logan, Utah 84322-0300*

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In crossed electric and magnetic fields the hydrogen atom undergoes a transition to chaotic scattering associated with a critical point in the Hamiltonian flow. The stability of the critical point is determined and leads to an accurate prediction of the transition to scattering that is independent of the magnetic-field strength. Nevertheless, observed variations in the apparent ionization threshold with magnetic-field strength are explained.

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In a landmark series of experiments, Raithel, Fauth, and Walther [1] have identified a class of quasi-Landau (QL) resonances in the spectra of rubidium Rydberg atoms in crossed electric and magnetic fields ($E\times B$). Similar to the original QL resonances observed by Garton and Tomkins [2], this set of resonances is associated with a rather small number of *planar* periodic orbits. In a different energy and field regime, Welge and co-workers [3] reported the first experiments on the hydrogen atom in crossed fields, also finding periodic orbits, as well as long-lived states lying far above the Stark saddle point. These they attributed to nonchaotic dynamics. Raithel and Walther [4] have further shown that the ionization threshold essentially follows classical scaling behavior over a wide range of field values, and pointed out that a maximum and minimum occur in the ionization threshold with varying magnetic field. In the same system, Main and Wunner [5] have demonstrated theoretically that, above threshold, the dynamics is scattering and chaotic [6]. These experimental and theoretical findings have far reaching implications for quantum physics [7] because of the insight they provide into the fundamental connection between classical and quantum mechanics [1,3,4,6,8].

In this Rapid Communication we provide a classical mechanism that explains and consolidates these interrelated findings for energies below, at, and above threshold. The atom is shown to undergo a transition to chaotic scattering related to the existence of a critical point in the Hamiltonian flow. This arises because of a velocity-dependent, Coriolis-like force in Newton's equations of motion. Velocity-dependent forces are common in many areas of physics, although the nonlinear dynamics of these systems is poorly understood. In addition, crossed fields arise in many diverse situations, including molecular problems, excitonic systems, plasmas, and neutron stars. The hydrogenic $E\times B$ problem therefore constitutes an experimentally accessible paradigm for a wide range of problems in atomic and molecular physics, solid state physics, nuclear physics, astrophysics, and celestial mechanics [6,9,10].

In scaled variables and cylindrical coordinates $(\rho,z,\phi)$ the $E\times B$ Hamiltonian is [1]

$$\mathscr{H}=\mathscr{E}=\frac{1}{2}\left(P_\rho^2+P_z^2+\frac{P_\phi^2}{\rho^2}\right)-\frac{1}{r}+\frac{P_\phi}{2}+\frac{\rho^2}{8}-\epsilon\rho\cos\phi, \tag{1}$$

where the scaled electric field $\epsilon=EB^{-4/3}$, and the scaled energy $\mathscr{E}=WB^{-2/3}$; $W$ and $B$ are the unscaled energy and magnetic field, respectively, and $r=\sqrt{\rho^2+z^2}$. At this point it is worth making a few general comments about the planar limit of Eq. (1), i.e., $z=P_z=0$: in the Zeeman limit ($\epsilon=0$) the system is integrable, as it is in the opposite, Stark limit when $B=0$ and $P_\phi=l_z$ is conserved. When the two fields are applied simultaneously both Eq. (1) and its planar limit are nonintegrable, although adiabatic invariants may exist [10,11].

It is often implied that the Stark saddle-point (SSP) criterion provides only a rough guide as to when direct ionization becomes energetically possible (e.g., $\epsilon\gtrsim 1.5$) [3,4]. This is because the SSP is based on an analysis of the potential energy in the $B=0$ limit. As $B$ is increased from zero the SSP criterion is, therefore, expected to deteriorate as a predictor of the ionization threshold. Nevertheless, the experimental ionization threshold was found to scale classically in Ref. [4]. This suggests that the transition from bound to scattering behavior might be related to the critical points of the Hamiltonian flow. In order to examine this possibility we determined the critical points of the flow generated by $\mathscr{H}$.

A remarkable result emerges from this analysis: there is a single critical point (for $\epsilon\neq 0$) that lies in the plane $z=0$ and occurs when $P_z=0$, $\rho=1/\sqrt{\epsilon}$, $\phi=0$, $P_\rho=0$, $P_\phi=l_z=-\rho^2/2$. The stability of the planar flow was determined by computing the eigenvalues of the matrix $L=JD^2H$ at the critical points [here, Hamilton's equations of motion are given by $\dot{\xi}=JDH$; $DH$ is the derivative of $H,\xi=(\mathbf{q}_i,\mathbf{p}_i)$, and $J$ is the symplectic form matrix [12]]. The four eigenvalues occur in pairs along the real and imaginary axes (i.e., $\pm\lambda_1,\pm i\lambda_2$ with $\lambda_{1,2}$ real) indicating that the motion at the saddle point is unstable. The critical point occurs at $\epsilon_{\rm crit}=(\mathscr{E}/2)^2$, which coincides precisely with the "approximate" SSP criterion, $\mathscr{E}=-2\sqrt{\epsilon}$. Thus, the SSP criterion

 

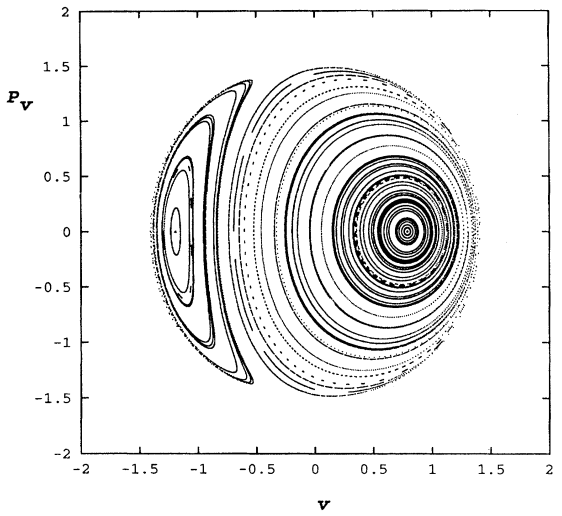


FIG. 1. Poincaré surface of section with $\mathscr{E}=-1.52$, $\epsilon=0.40$.

should hold for *any* value of $\epsilon$ irrespective of the absolute size of the magnetic field $B$. This finding is true only in the infinite nuclear mass approximation and results from a cancellation of terms in the derivative $H_\rho$. Situations might exist in other $E\times B$ Rydberg problems, e.g., in exciton models, where the coefficients of these terms do not lead to such a cancellation [10], and the critical point analysis may be expected to reveal further complexity. This is best analyzed using the concept of a zero velocity surface [13] as has been done recently in the related problem of a hydrogen atom subjected to a circularly polarized microwave field [14]. We note that this result agrees with the analysis of Clark *et al.* [15,16] who further found that the Stark saddle point could not be stabilized by addition of a magnetic field.

To investigate our findings numerically, we specialized to the planar system for which Hamilton's equations of motion are most easily integrated in regularizing parabolic coordinates $(u,v)$ with $x=(u^2-v^2)/2, y=uv$ [17]. The governing Hamiltonian becomes

$$\mathscr{K}=\frac{2}{\Omega}=\tfrac{1}{2}(P_u^2+P_v^2)+\tfrac{1}{2}(u^2+v^2)-\frac{\epsilon}{2\Omega^3}(u^4-v^4)+\frac{1}{4\Omega^2}(u^2+v^2)(uP_v-vP_u)+\frac{1}{32\Omega^4}(u^2+v^2)^3, \tag{2}$$

where $\Omega=\sqrt{-2\mathscr{E}}$. The dynamics were examined by computing Poincaré surfaces of section defined by $u=0$ and selecting the branch of $P_u$ that corresponds to taking the positive branch of the square root. The motion was studied at fixed $\mathscr{E}$ while varying $\epsilon$, although one could, alternatively, fix $\epsilon$ and vary $\mathscr{E}$.

Figure 1 is a compilation of surfaces of section for a randomly picked set of trajectories with $\epsilon=0.4$ and $\mathscr{E}=-1.52$. For this value of $\epsilon$ the dynamics is regular and consists of two islands at whose centers are located stable fixed points. In the limit $\epsilon=0$ the islands are symmetric about the origin and reflect orbits whose angular momenta are in opposite senses with regard to their rotation about the

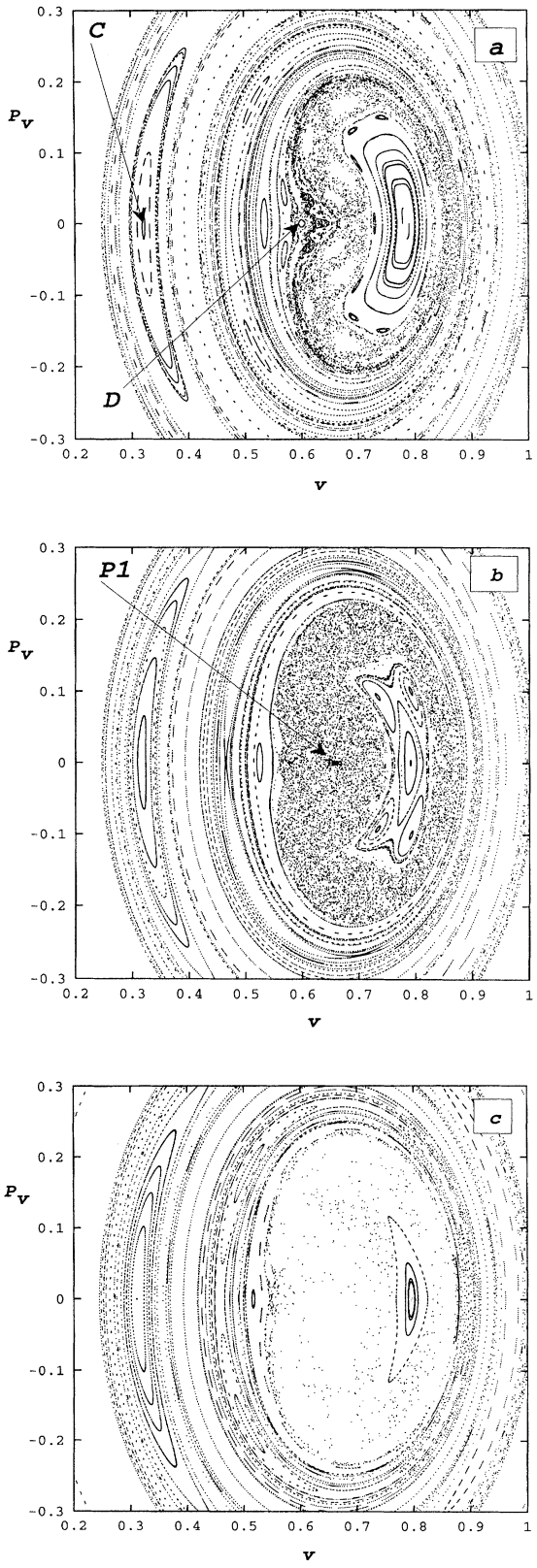


FIG. 2. Enlargements of Poincaré surfaces of section with $\mathscr{E}=-1.52$ and (a) $\epsilon=0.5765$, (b) $\epsilon=0.577\,598$, and (c) $\epsilon=0.5785$.

origin [10]. The left- (right-) hand island corresponds to positive (negative) values of $l_z$. The fixed points then correspond to stable, high-angular-momentum, circular periodic orbits.

As $\epsilon$ is increased beyond 0.4 the right-hand ($l_z<0$) island suddenly gives birth to a chaotic region that surrounds the original fixed point. Figure 2 is a series of enlargements of the chaotic region for values of $\epsilon$ bracketing the transition that occurs at $\epsilon=0.5776$. The label $C$ on Fig. 2(a) indicates the position of one of the principal periodic orbits contributing to the spectra in Refs. [1,18]. Over an extremely small range of $\epsilon$ the dynamics first becomes chaotic and is accompanied by a marked proliferation of periodic orbits. In Fig. 2(b) the periodic orbit labeled $P1$ is on the verge of experiencing a tangent bifurcation—the eigenvalues ($\lambda_\pm$) of the Jacobian for the two-dimensional symplectic map collide at $\lambda_\pm=1$ and then split off along the positive real axis [19,20]. After the system has passed through this bifurcation the chaotic region disappears by turning into a scattering regime visible as a "hole" in Fig. 2(c) in which few or no points appear—eventually the region disappears completely, even the large island on the eastern edge of the region in Fig. 2(c) disappears, leaving only an expanding hole as $\epsilon$ is increased further.

Many of the periodic orbits visible in Fig. 2(a) resemble the $C$-type orbits of Ref. [1] that were shown to contribute strongly to spectra; i.e., they undergo close encounters with the nucleus [21]. Our calculations suggest that for the parameter values reported in Ref. [1], i.e., $\epsilon=0.68$ and $\mathscr{E}=-1.50-0.034\,N$, $N=0,1,2,\ldots,6$, a proliferation of periodic orbits, followed by a transition to chaotic motion and scattering will occur for $N_{\rm crit}\approx 4$. Of course, the experiments relate to Rb, while the calculations are for the H atom, so conclusions must be drawn cautiously. However, careful examination of the Fourier spectra in Ref. [1] indeed reveals that a transient regime exists in which the number of QL resonances increases while, at the same time, the spectra become noticeably grassier, suggesting that the dynamics is at least partially chaotic. This transition occurs in precisely the range ($N\sim 3-4$) predicted by Hamiltonian stability theory. Interestingly, the spectrum in Ref. [1] with $N=0$ is well above the ionization threshold, but appears regular. Effectively, many of the periodic orbits and the chaotic region itself have been obliterated since they can now escape over the saddle; thus any experiment that is sensitive to bound dynamics will result in extremely regular-looking spectra that are dominated by the small number of periodic orbits that manage to survive the sudden disappearance of the chaotic region. This is in complete agreement with the experimental findings of Wiebusch *et al.* [3] that long-lived, nonchaotic states exist far above the Stark saddle point.

Of course, strictly speaking, this is not an authentic scattering system because there is no flux of electrons coming in from infinity. Initially, the electron is located close to the nucleus and, upon excitation, the system is, perhaps, best thought of as a half-collision. We computed the time delay (residence time in the chaotic region) as a function of the initial coordinate $v$, restricting initial conditions to the surface of section as defined earlier, and find that the time delay function is self-similar [6,22,23]. This is consistent with the simulations of Main and Wunner [5]. The present analysis shows clearly that the observed self-similarity is a direct result of the system reaching a critical point when already chaotic orbits are suddenly destabilized; compare Figs. 2(b) and 2(c).

It remains to explain the observation in Ref. [4] that the ionization threshold is a maximum, $I_{\rm max}$, when $\epsilon=0.058$, $\mathscr{E}=-0.45$; and a minimum, $I_{\rm min}$, when $\epsilon=0.19$, $\mathscr{E}=-0.74$. The critical-point analysis leads us to argue that the classical ionization threshold should be independent of the individual values of $\epsilon$ and $\mathscr{E}$. In the classical simulations and in the experiment reported in Ref. [4] the ionization threshold is defined with reference to the fraction of atoms (e.g., 50%) that ionize. Not all trajectories, however, have access to the ionization channel. Many orbits, even above threshold, are separated from the chaotic region boundary of scattering orbits by intact Kolmogorov-Arnold-Moser (KAM) tori as shown in Fig. 2(c). The mixed nature of phase space thus suggests that the *observed* ionization threshold will depend strongly on the relative size of the boundary as compared to the rest of phase space. Computation of surfaces of section corresponding to $I_{\rm max}$ and $I_{\rm min}$ reveals that, in both cases the scattering boundary exists; i.e., the system is above the ionization threshold. However, the volume of phase space for which scattering is possible is substantially smaller at $I_{\rm max}$ than at $I_{\rm min}$. Note that at $I_{\rm min}$, $\epsilon$ is $\approx 140\%$ of the threshold value, while at $I_{\rm max}$, $\epsilon$ is only $\approx 114\%$ of threshold. This leads to the observed difference in the *apparent* ionization thresholds, i.e., the ionization probabilities differ but the ionization threshold is the same.

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