Computation of the Arnol'd Web for the Hydrogen Atom in Crossed Electric and Magnetic Fields

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A Rydberg atom placed in crossed static electric and magnetic fields is presented as a new testbed for phenomena not possible in two degrees of freedom. We compute the Arnol'd web for this system and explore the time scale and the physical consequences of diffusion along this web.

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Resonances between various motions in multidimensional systems (be they galaxies [1], the solar system [2], or particle accelerators [3]) determine much of the character of the dynamics. A striking confirmation of the universal role of resonances is provided by the KAM theorem [4]. In the last decade evidence has been accumulating that quantal effects are also anchored in subtle ways to the classical dynamics in and near nonlinear resonances, the microwave ionization of excited hydrogen atoms being a well-researched case in point [5]. These findings establish the resonance structure of a dynamical system as one of the most worthwhile goals in analyzing the dynamics. However, for realistic multidimensional systems, this task is far from trivial and often founders on identifying the proper action-angle variables which describe the nonseparable system in question. The purpose of this Letter is to report on a successful computation of the network of resonances (the so-called Arnol'd web) of a highly perturbed atomic system with three degrees of freedom, a Rydberg atom placed in two crossed static external fields [6,7]. After exploring directions, rates, and physical consequences of multidimensional diffusion, we show that in one specific direction it leads to drastic changes in atomic properties and, in particular, to a flattening of the atom. To our knowledge, this is the first such calculation in atomic physics.

We also provide an answer to a long-standing question in dynamics: Can Arnol'd diffusion ever be fast enough to be detected in a realistic physical system? In some accordance with conventional wisdom, we find that even the fastest physical manifestations of Arnol'd diffusion require thousands of Kepler periods to become noticeable. Of course, these long evolution times would normally preclude direct detection, but recent discoveries concerning nondispersive electronic wave packets in a related class of problems [8,9] have altered this unfavorable outlook. The crossed fields problem of this Letter is fundamental since insights gleaned from this system (such as critical point analysis [10] and periodic orbits [6,7]) have led to the study of related but more intricate scenarios such as Rydberg atoms in circularly polarized (CP) fields [8,11] and the CP problem with a stabilizing magnetic field [12], for which nondispersive electronic wave packets have been shown to exist. Again, the crossed fields problem provides the simplest and cleanest example of Arnol'd diffusion within this class of systems, and it is reasonable, as our preliminary results show, that the time scales involved are consistent with the very long lifetimes of these nondispersive wave packets.

Furthermore, our chosen system represents a departure from the one and two degree of freedom systems that dominate Rydberg atom physics. The classical dynamics of multidimensional systems undergoes a fundamental change when the number of degrees of freedom exceeds two: Upon crossing that threshold a wealth of new physics becomes possible [13]. Yet forays into this vast area are comparatively rare because beyond this divide we lack analytical tools comparable in power to the Poincaré surface of section, and newly developed computational methods, such as the one explained below, offer the only viable hope of analyzing the new physics. The stark contrast between systems possessing two degrees of freedom and those with more can perhaps be best illustrated in the space of actions of integrable and nearly integrable systems. In a conservative system with two degrees of freedom invariant tori form impenetrable obstacles to free movement and only diffusion across the stochastic layers is important. However, for systems with more than two degrees of freedom, stochastic layers merge to form the Arnol'd web and any surviving tori can no longer impede diffusion. Diffusion becomes much richer now, and, in particular, it allows for the possibility of large scale diffusion along resonances, a diffusion process generally associated with Arnol'd [4,13]. Stochastic motion might carry the system arbitrarily close to any point on the energy shell by traveling along the resonance layers and simply circumventing invariant tori. Usually, diffusion across a stochastic layer, which is generic to Hamiltonian systems with more than one degree of freedom, is much stronger than diffusion along a resonance, although under certain conditions the rate can be appreciable [14]. Of course, in a realistic system such as the crossed fields problem, both kinds of diffusion occur, the stronger usually drowning out the weaker.

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The perturbed Coulomb Hamiltonian for crossed electric and magnetic fields is given in atomic units by [15]

$$\mathrm{H} = \frac{1}{2}\,p^2 - \frac{1}{r} + \frac{B}{2}\,L_z + \frac{B^2}{8}\,(x^2 + y^2) + Fx\,. \tag{1}$$

Here, the magnetic field B is in units of 2.35×10^5 T and points in the z direction, the electric field F is in units of 5.14×10^{11} V/m and points in the x direction, and L_z is the z component of the angular momentum $\mathbf{L} = \mathbf{r} \times \mathbf{p}$. The symmetry breaking of the Coulomb potential [16] induced by these crossed fields leaves no continuous symmetry intact and provides us with a system the full dynamics of which unfolds in three degrees of freedom. This atomic system combines several major advantages: It can be studied in the laboratory with superior resolution [6], and, as an additional twist, is quantum mechanical while at the same time being in the correspondence principle regime where reasonable agreement with classical mechanics can be expected. Indeed, such atoms have long been recognized as atomic-scale laboratories where the quantum mechanics of highly nonlinear systems can be tested. Long-time, high-accuracy trajectories can be computed using Meyer's algorithm [17] after regularizing the Coulomb singularity in Kustaanheimo-Stiefel coordinates [18]. Our results can then be transferred to weak laboratory fields and high values of the principal quantum number n (where semiclassical mechanics is valid), using the scaling properties of the system [19].

At constant energy, the action space can be displayed easily since it is two dimensional. However, it is far from straightforward to compute these actions without knowing the shape of the tori. We used two powerful methods to characterize invariant tori or their remnants, and thus to locate the electron's orbit in action space. Assuming ergodicity on the torus, the location is found by analyzing the properties of a single trajectory on the torus, and the change of these properties with time represents a measure of the diffusion strength. The first of these methods, Laskar's frequency analysis (LFA) [20], relies on the motion being quasiperiodic on an invariant torus with fundamental frequencies $\mathbf{f}$ such that a dynamical variable g can be expressed as

$$g(t) = \sum_{\mathbf{m}^{(k)}} c_k e^{i(\mathbf{m}^{(k)},\mathbf{f})t}, \tag{2}$$

with complex coefficients c_k and integer vectors $\mathbf{m}^{(k)}$. Sampling at consecutive time steps, LFA yields very accurate approximations to the most important frequency combinations $\mathcal{F}_k = (\mathbf{m}^{(k)}, \mathbf{f})$ and the associated coefficients c_k. For broken tori, the quasiperiodicity is fulfilled only approximately, yet the method often manages to give useful results. However, unless great care is taken in the choice of the appropriate variables, the most important frequencies may change across action space, thereby leading to different linear combinations. In our experience, LFA is very accurate for the determination of local diffusion, revealing even the weakest resonances. Alternatively, tori may be identified by the time averages of N linearly independent dynamical variables

$$\langle g \rangle_t = \int_t^{t+T} g(t')\,dt' \tag{3}$$

for sufficiently long time intervals T. While not as accurate as LFA for a given integration time, this method is simpler and gives stable results even along broad chaotic layers, making it the method of choice when diffusion is extensive. In such a scenario, the accuracy in determining the position in action space is naturally limited by diffusion itself.

Figure 1 shows, on a two-dimensional grid of initial conditions, the local diffusion strength defined as $\sqrt{(\Delta\mathcal{F}_1)^2 + (\Delta\mathcal{F}_2)^2}$, where Δ means the difference between the time intervals $[0, T]$ and $[T, 2T]$ ($T = 600$ Kepler periods T_{Kepler}) for the two most important frequency combinations $\mathcal{F}_1, \mathcal{F}_2$ of the intermediate motion (see below) computed with LFA. The Arnol'd web of resonances is clearly visible as a network of bright lines with strong diffusion. At the crossings of resonances [where two independent resonance relations $(\mathbf{m}, \mathbf{f}) = 0$ are fulfilled] the motion becomes periodic or near periodic. By plotting ensemble averages $\langle n \rangle$ vs $\langle L_x^2 \rangle$ on the same grid (Fig. 2), we obtain a map of action space at fixed energy. In this figure as in Fig. 1, each point is shaded according to diffusion strength. Resonances reveal themselves by distorting the regular grid of initial conditions [21]. Diffusion in action space can now be tracked by plotting the values of $(\langle L_x^2 \rangle, \langle n \rangle)$ at consecutive time steps.

In Fig. 2 we display examples for both types of diffusion mentioned in the introduction. Classical perturbation theory applied to this system shows that the motion of the electron is composed of three motions of differing time scales: a fast motion along Kepler ellipses, an intermediate evolution of the elements of these Kepler ellipses (linear in the fields), and a slow motion along the contour lines of an adiabatic invariant (quadratic in the fields) [19]. A trajectory starting at point (1) in Figs. 1 and 2, which is inside the 1:0:4 resonance between these three motions, merely bounces from edge to edge across the chaotic layer. This is the "across" diffusion [13] which is also found in lower dimensional systems. It is usually rather strong, as is the diffusion in the vicinity of resonance crossings, while the diffusion along resonances is usually much weaker and indeed not noticeable in the regular (dark) regions on the time scales considered here.

In contrast, a trajectory which is launched within the thin (1:2:3) resonance layer at the sign of the cross [(2) in Figs. 1 and 2] diffuses along this resonance for $10^5 T_{\mathrm{Kepler}}$ until it escapes into the broad intersection of resonances [marked (3)]. It is temporarily trapped there for a long time ($5 \times 10^6 T_{\mathrm{Kepler}}$). This trapping could also be detected at the other strong resonance crossings, albeit in a much weaker form. The trajectory then escapes

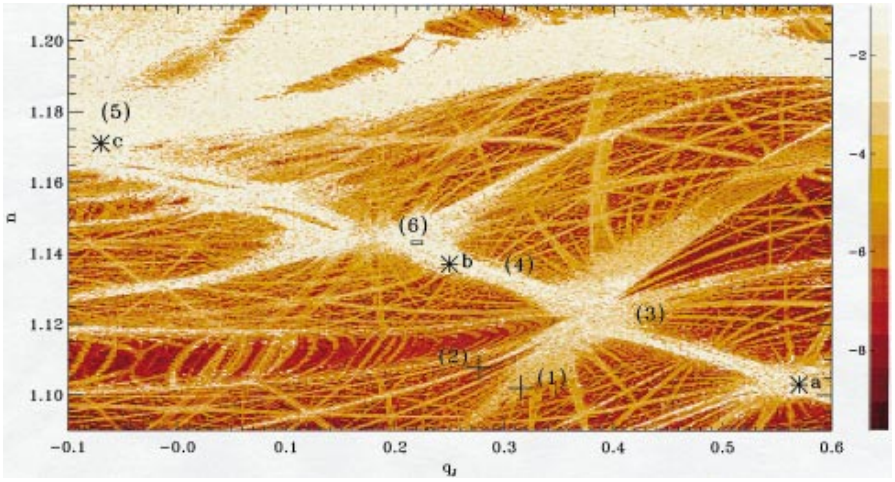

FIG. 1(color). Local diffusion strength as defined in the text on a logarithmic scale as a function of initial conditions. Bright colors mean strong diffusion. The fields are $B = 0.40$, $F = 0.06$. The energy is fixed at $E = -0.5$. The variable n on the y axis is the classical analog to the principal quantum number, $n^2 = \mathbf{L}^2 + \mathbf{A}^2$, where $\mathbf{L}$ is the angular momentum of the electron and $\mathbf{A}$ the Runge-Lenz-Laplace vector [18]. The variable q_J is defined as $q_J = 2J_x/\sqrt{n - 2J_z}$ where $\mathbf{J} = (\mathbf{L} + \mathbf{A})/2$ [19]. (Available at http://www.gatech.edu/physics/arnold/arnold.html.)

very quickly (within less than $2 \times 10^4 T_{\text{Kepler}}$) along the strong central (1:4:5) resonance [marked (4)] to the large chaotic region (5) in the upper left-hand corner. Remarkably, it returns several times into the central resonance and even close to its birthplace (not shown in figures). Typically, trajectories in the large chaotic region would repeatedly enter regular regions characterized by a small local Lyapunov exponent. This shuttling between the chaotic zone and the regular areas constitutes a variety of intermittency (see also [15]).

The diffusion along the central resonance discussed above is the fastest "along" diffusion that we found in this system. We define a diffusion coefficient in terms of the ensemble average of the principal quantum number n as

$$D(t) = \frac{(\langle n\rangle_t - \langle n\rangle_0)^2}{2t}, \tag{4}$$

and average it over a random ensemble of trajectories originating from the box (6) of Fig. 1. Its value is around $2 \times 10^{-7} T_{\text{Kepler}}^{-1}$ and on the average it is directed towards regions of higher diffusion at larger values of $\langle n\rangle$ and in the large chaotic zone, so that the diffusion coefficient increases slowly in time. After $8000 T_{\text{Kepler}}$, 5% of the trajectories have escaped from the box into the chaotic zone.

The diffusion towards higher values of $\langle n\rangle$ has direct physical consequences. A trajectory diffusing along the strong resonance drastically changes its shape and properties (see Fig. 3). The most striking feature is the flattening of the atom; it gradually adopts the topology of the most stable periodic orbits which are planar [6]. The increase in $\langle n\rangle$ is strongly correlated with an increase in the average distance $\langle r\rangle$ from the core and the distance of closest approach to the nucleus. The latter increases by 40% as a result of diffusion. This flattening is also associated with

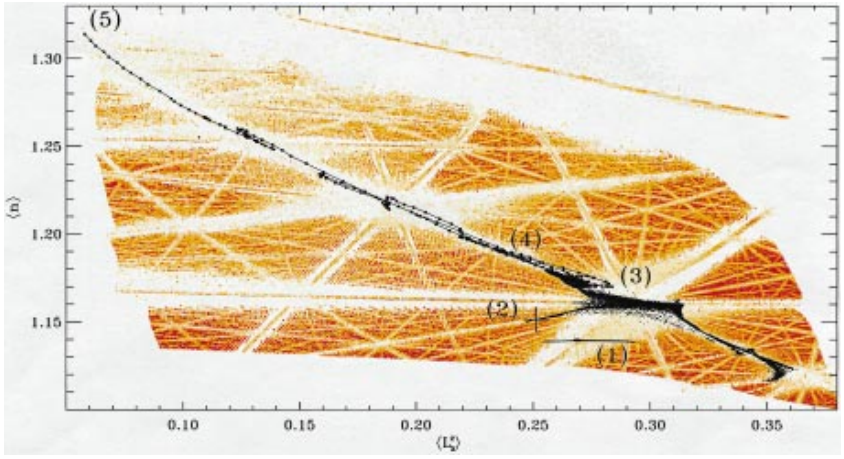

FIG. 2(color). The Arnol'd web plotted as $\langle n\rangle$ vs $\langle L_x^2\rangle$. Two examples of trajectories wandering through the web are shown (see text). The knots along the trajectories occur at time steps of about $300 T_{\text{Kepler}}$. The points are connected along the large resonance only. (Available at http://www.gatech.edu/physics/arnold/arnold.html.)

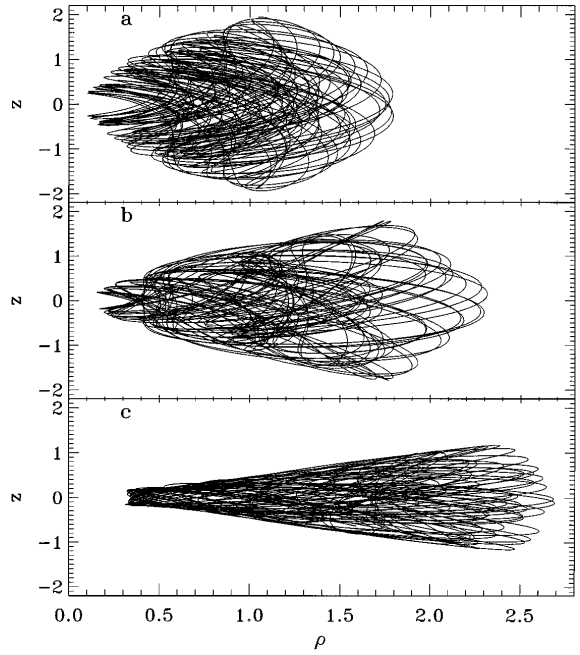

FIG. 3. Trajectories are launched at three places along the strong resonance (marked by stars in Fig. 1). We show the change in character as the electron diffuses along the resonance by plotting z vs $\rho = \sqrt{x^2 + y^2}$. Note the flattening of the atom.

a 70% increase in the dipole moment (which, however, remains small). These changes have a rather drastic impact on the ionization probability of the system as the electron may only ionize during close encounters with the core.

In conclusion, we have identified and analyzed the multidimensional resonances of an experimentally accessible atomic system that may well form a paradigm for phenomena in several degrees of freedom. We calculated its Arnol'd web and contrasted the across and along diffusion. The diffusion along the central resonance in Fig. 2 might affect the quantum dynamics noticeably, and the newly discovered long-lived, nondispersive electronic wave packets [8] might be used for a comparison of these classical results with quantum-mechanical ones.

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