Intramanifold Chaos in Rydberg Atoms in External Fields

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Highly excited Rydberg atoms are atomic-scale laboratories where the quantum mechanics of chaotic systems can be tested. The extensive symmetry breaking introduced into the Coulomb potential by crossed electric and magnetic fields removes the obstacles for phenomena not possible in 2 degrees of freedom. We bring out the classical structures that support the complexity of motion and lead to alternating layers of order and chaos. The special status assigned to certain dominant periodic orbits by experiment emerges naturally in our treatment.

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The purpose of this Letter is to report on some remarkable classical structures that support the great complexity of electronic motion in Rydberg atoms in crossed external fields. Confronting classical and quantum mechanics in regimes where the classical motion is chaotic is one of the fundamental problems of this decade, as evidenced by the enormous outpouring of research in the subject. In particular, the attention of nonlinear dynamicists and atomic physicists has become focused on electron dynamics and spectroscopy of highly excited Rydberg atoms placed in external fields. There are two reasons for this intense interest: These atoms can be prepared and manipulated in the laboratory, and they are amenable to theoretical treatment, often with astonishing accuracy [1,2]. Their Hamiltonians possess simple nonlinearities which lead to chaotic dynamics. The very high excitations involved imply that detailed quantum treatment can be tedious; on the other hand, the same high excitations place the electron in a regime where reasonable accuracy can be expected of the correspondence principle. Therefore, Rydberg atoms in static external fields constitute atomic-scale laboratories where the quantum mechanics of highly nonlinear systems can be tested [3].

The symmetry breaking of the Coulomb potential [4] induced by external fields (in practice the magnetic field is of greater interest since the Stark problem is separable) affects the three quantum numbers n, l, m_l of the Rydberg electron differently: As long as a single field direction is present, m_l remains a good quantum number, l breaks down extensively, whereas n breaks down only gradually with increasing magnetic field [5]. Therefore, an n manifold of electronic energy levels does not have enough degrees of freedom for chaos, which only develops when different n shells mix ("intermanifold chaos"), usually close to the ionization threshold. In contrast, the extensive symmetry breaking introduced by two misaligned (in practice, crossed) fields leaves no continuous symmetry intact and thereby opens the floodgates for a wealth of new physics which is only possible beyond 2 degrees of freedom: For instance, Arnol'd diffusion can take place in this experimentally accessible system. Less esoterically, it also becomes possible to excite wave packets localized in all spatial dimensions, and the observation of these wave packets, which was accomplished recently [6], opens an exciting window on the dynamics of the electron. Yet another is "intramanifold chaos": the minimum number of dimensions required for chaos is reached within an n shell of the atom, often well before the ionization threshold. The easy accessibility of the chaotic region and the prospect of tuning the extent of chaos through external means offer great advantages for experimental investigations.

The deceptive simplicity of the perturbed Coulomb Hamiltonian (in atomic units)

$$\mathcal{H} = \frac{1}{2} p^2 + \frac{B}{2} m + \frac{B^2}{8} (x^2 + y^2) - \frac{1}{r} + Fx \quad (1)$$

is belied by the rich nonlinear dynamics it generates. Here, the magnetic field B points in the z direction, the electric field F is in the x direction, and m is the z component of the electronic angular momentum $\mathbf{L} = \mathbf{r} \times \mathbf{p}$. Great complexity is evident in the recent high-resolution photoabsorption experiments of the Welge [7] and Walther [8,9] groups whose successes in relating some of their peaks to periodic orbits is evidence that the periodic motions of the electron underlie much of the classical-like oscillations in these spectra, a finding which constitutes an elegant experimental connection between classical and quantal theories, as well as being a striking demonstration of the Gutzwiller formalism [10]. To identify the classical structures that support these complex spectra is a great challenge. Given the Hamiltonian, it might be thought straightforward (if tedious) to perform a systematic search for periodic motions, until one realizes the magnitude of the task: This Hamiltonian has an enormous number of recurring and periodic motions [11] and even equipped with the various field- and quantum-number dependent broken symmetry chains of the SO(4) group, this program will amount to little more than searching for a needle in a haystack.

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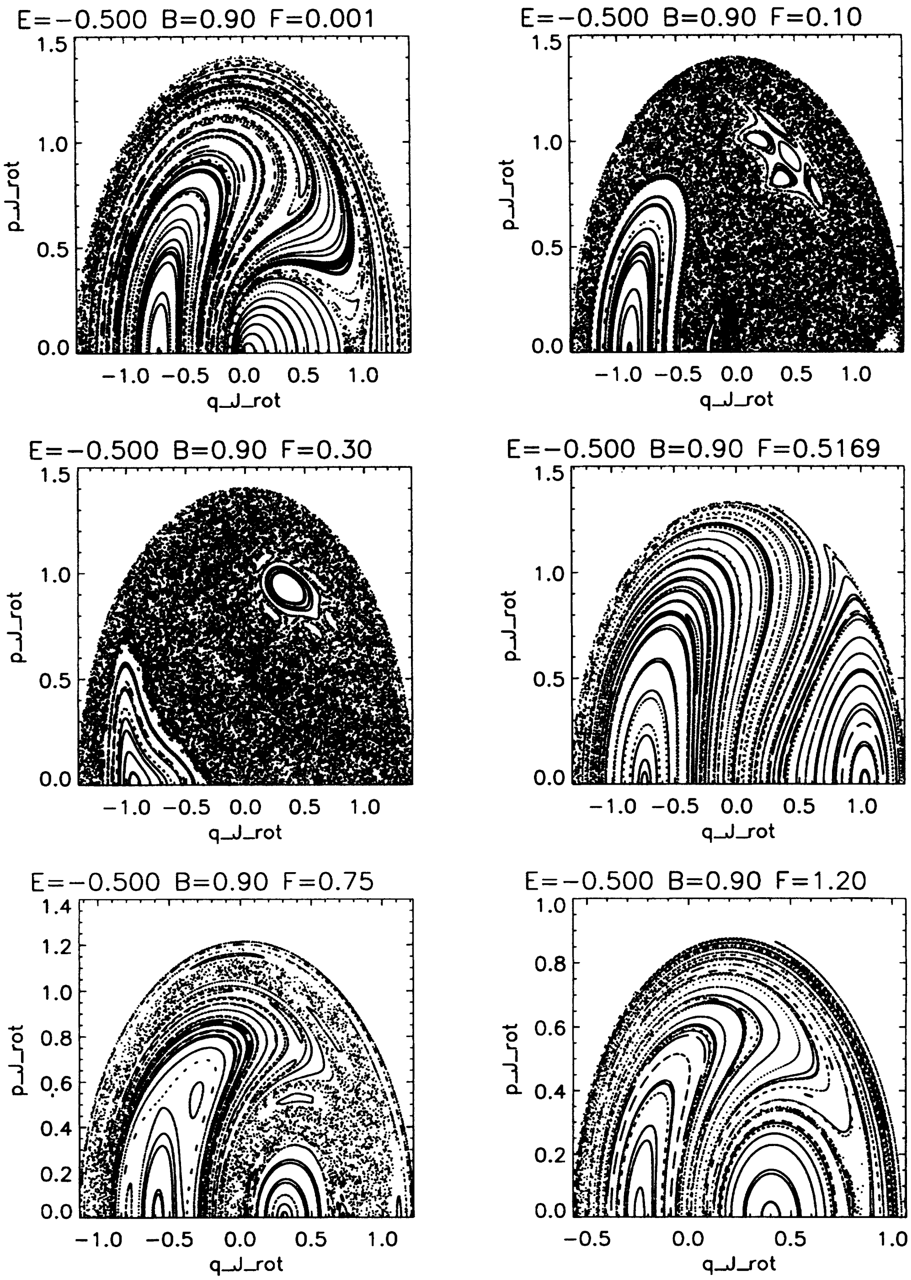

FIG. 1. Poincaré surfaces of section (PSOS) generated from the approximate Hamiltonian. The magnetic field B is kept constant, and the electric field is increased steadily. Note the stable fixed point on the left hand side, which is robust throughout these changes of parameters. Chaos seems to originate from the unstable fixed point on the right. These two motions differ from each other solely through the relative phases of the J and K rotors (see text).

In this Letter, we demonstrate how this intricate and important problem can be simplified considerably by expressing it in terms of coupled asymmetric tops. The resulting approximate Hamiltonian reveals not only the experimentally significant periodic orbits and their bifurcation behavior but also some intriguing order-to-chaos alternations, presumably induced by novel adiabatic invariants. The technically involve derivation [12] is as follows. The regularization [10] of Hamiltonian (1) in the four Kustaanheimo-Stiefel coordinates u [13] and their conjugate momenta p_u leads to a pseudo Hamiltonian which is then analyzed using the succession of canonical transformations of the F_2 type of canonical perturbation theory [14]. When the enormous number of terms in the resulting Birkhoff normal form are collated in terms of the two angular momenta $\mathbf{J}$ and $\mathbf{K}$ inherent to the Coulomb problem [15], the Lie algebraic generators of the group SU(2)⊗SU(2) [locally isomorphic to the symmetry group

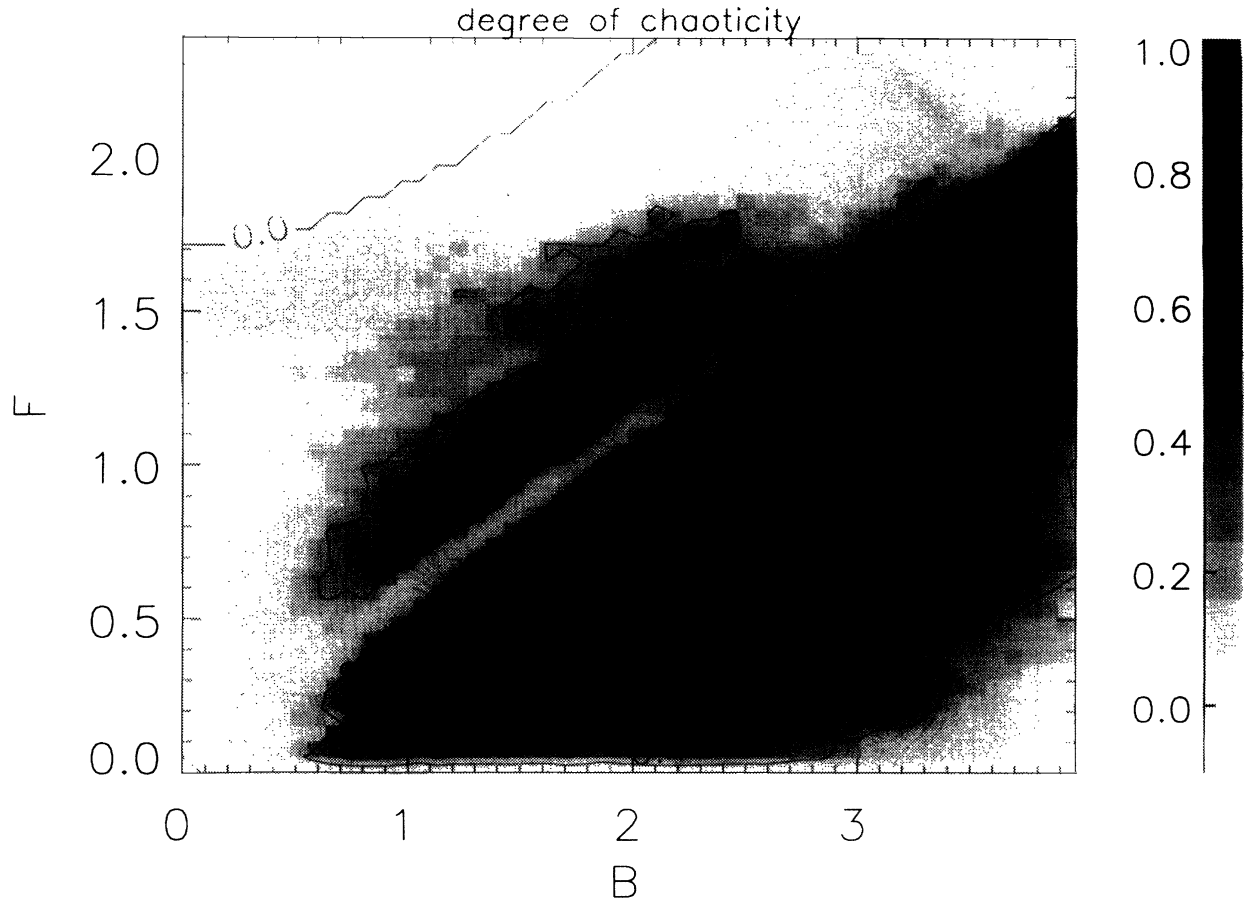

FIG. 2. Degree of chaoticity of the phase space: The percentage of chaotic trajectories with initial conditions $p_J = p_K = 0$, which by symmetry are the most significant set of initial conditions. Note the "ridge of stability" along the line $F/B = 4/7$. Clearly, by changing the B and F fields judiciously, the system can be made to undergo repeated transitions between chaos and order.

SO(4) of the Coulomb problem], the normal form, correct to second order in the fields, contains a pair of degenerate, strongly coupled asymmetric tops in **J**, **K**. The action-angle forms of these generators which are needed for subsequent classical-mechanical analysis are best expressed in terms of the extended Lissajous variables [16,17]. Because n, which determines the norm of **J** and **K**, is a conserved quantity in our treatment, this normalization reduces the problem to 2 degrees of freedom. We performed our classical calculations at $n = 1$, though any other n will do as long as the fields are scaled accordingly. The dynamics is governed by the quantities Bn^3 and Fn^4 for which we adopted experimentally realistic values.

The Poincaré surfaces of section (PSOS), when displayed in terms of the conjugate dynamical variables of the rotors, show an order-chaos-order transition as a function of increasing electric field (Fig. 1). While one expects a strongly coupled nonlinear system to show a transition from order to chaos with increasing coupling, its return to order with yet higher coupling is unusual and attests to the intriguing complexity of the system. These PSOS's are generic for many other scenarios of the crossed fields system; for instance in cases where both fields are increased simultaneously (see below). The PSOS's show further that one stable (on the left hand side) and one unstable (on the right hand side) motion result from the coupling of these two unstable rotations. Chaos spreads from the unstable fixed point on the right. This alternation can be understood in terms of the rotor picture. Each of the two asymmetric tops in that Hamiltonian has one unstable and two stable axes of rotation. One intuitively expects erratic motion when both tops are rotating around their unstable intermediate axes. Indeed, the parameter range where the overall motion is most chaotic is not far from this double-unstable excitation of the pair of tops as can be verified by studying the exact dynamics. Of course, their strong (bilinear) coupling alters this idealized picture of the dynamics: The Poincaré SOS's in Fig. 1 show that of the two motions resulting from the coupling of two asymmetric top rotations, one is stable and robust to changes of external parameters, whereas the other (the one on the right hand side) is not. However, when viewed in the asymmetric top variables, these radically different motions are distinguished only by the relative phase of the two rotors in **J** and **K**: In the stable one, the phase difference is π, whereas in the unstable one the rotors are in phase.

A striking connection to laboratory Rydberg spectra is established by examining the fixed points of the PSOS. When the motion contained in the robust stable fixed point is translated to Cartesian coordinates, it turns out to be the one planar periodic orbit (denoted C_1 by Raithel, Fauth, and Walther [9]) which dominates most of their recent photoabsorption spectra. In our asymmetric top description, its prominence is clearly connected to the stability of the motion, and its special status is evident at once (it is worth recalling that our model does not presuppose planarity of motion). Of course there are other fixed points surrounding this one, and, when they are examined, they all turn out to correspond to other prominent periodic orbits of the system (often of astonishing complexity) implicated by experiments differing in their laser polarizations [8]. Further windows on the dynamics are opened by examining the percentage of chaotic trajectories as a function of electric and magnetic fields (Fig. 2). Repeated transitions between order and chaos are observed, when, for example, the electric field is increased at constant magnetic field (indeed, the PSOS's in Fig. 1 correspond to various electric fields at magnetic field $B = 0.9$ atomic units in this diagram). This layering indicates that the complex phase space contains stable classical structures within chaotic areas. One of these structures becomes evident when the electric and magnetic fields are increased in a roughly 4:7 ratio: The system walks on a ridge of stability deep into the chaotic region, indicating that for these parameter values there is an adiabatic invariant [belonging to the subgroup $O(3)_Q$ [11]].

Given the enormous size of the parameter and phase space of the problem, our treatment of the dynamics is a considerable advance since the location and nature of these special areas and motions would be very difficult to pinpoint without the guidance of our picture. In particular, the special status assigned to some intricate periodic motions of the electron by recent experiments is connected naturally to their stability properties through our treatment. Partial support of this research by the A. von Humboldt Foundation is gratefully acknowledged.

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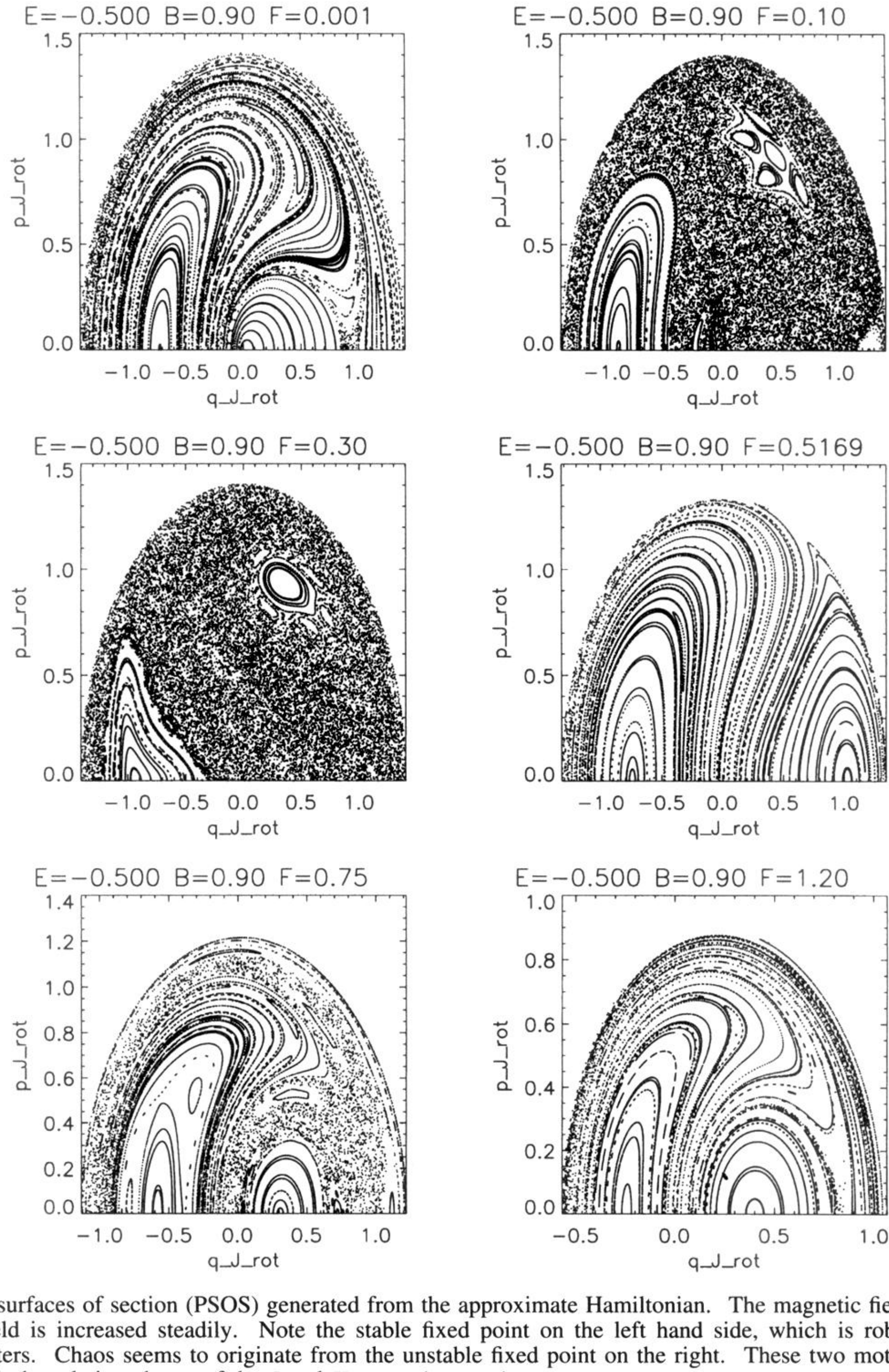

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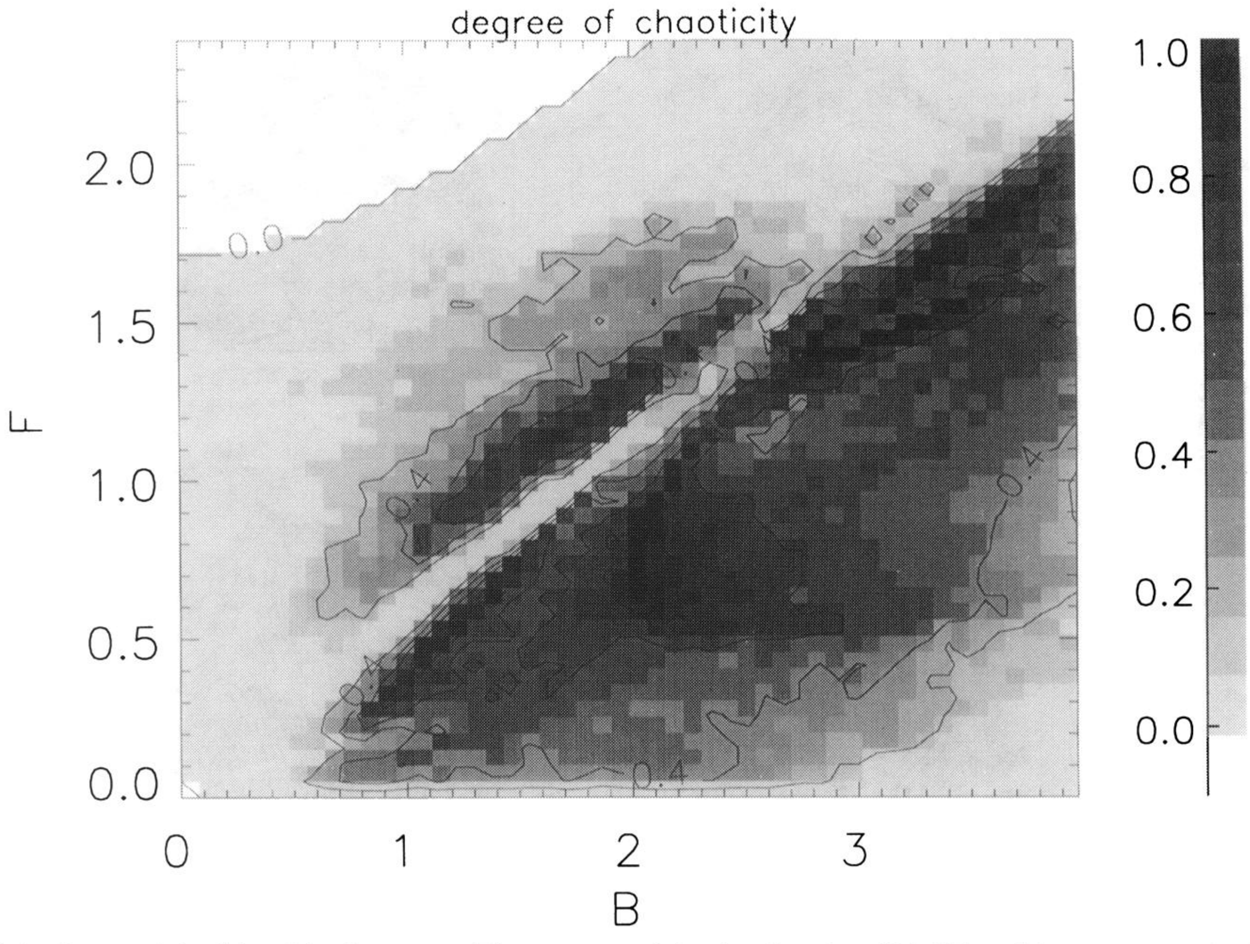

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