Incidentally, we appear to have here, for the first time, an objective way of distinguishing 'active' from 'passive' motion, a concept that was introduced by Townsend[18]. The very simple criterion we have found is that low fluctuations (typically less than a standard deviation in the velocity components) contribute little to the flux, and could perhaps be identified with passive motion; the more intense fluctuations are 'active' in the generation of flux. An attractive possibility is that the passive motion is best described in the language of waves, whereas the active motions – productive or counter-productive – are best seen as a series of events. So I suggest a tentative answer to the question in the title that turbulence can be both waves and events – but the waves are passive, and all the flux comes from the events, which (to a first approximation) are always members of a signed two-parameter family, with the positive events outnumbering and outlasting the negative ones in a neutrally stable flow.

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Rydberg atoms and molecules – Testing grounds for quantum manifestations of chaos

M. Lakshmanan and K. Ganesan

Understanding quantum manifestations of classical chaos is of intrinsic and practical interest. In the recent years several fingerprints of chaos have been recognized in microscopic systems. In this connection Rydberg atoms and molecules, which are highly excited systems under various external interactions, are found to be testing grounds to understand quantum chaos. We trace here these recent developments and discuss their implications and future outlook.

THE deterministic randomness or chaos exhibited by generic nonlinear dynamical systems has been found to present significant practical and philosophical implications[1–5], and probably limitations[6] as well, in the quantum description of microscopic world. There is no doubt that quantum theory is a more accurate description of nature. However, Bohr's correspondence principle requires that in the appropriate limit the remnance of the signatures of (classical) chaos (of macroscopic world), namely the exponential divergence of nearby trajectories and the intrinsic uncertainty due to nonlinearity, should follow, barring unforeseen singularities in the Planck

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constant $\hbar \rightarrow 0$ limit which possibly might prevent the smooth transition from quantum mechanics to classical mechanics[6]. Search for such quantum manifestations of classical chaos in the practical sense, which goes by the terminology 'quantum chaos' or 'quantum chaology', has recently attracted considerable interest[1-5]. For example, one might look for possible fingerprints of chaos in the eigenvalue spectrum, wavefunction patterns and so on.

In recent times it has been found that highly excited Rydberg atoms and molecules (which are effectively one-electron systems) under various external fields[7-14] are veritable goldmines for exploring the quantum aspects of chaos. These systems are particularly appealing as they are not merely mathematical models but important physical systems which can be realized in the laboratory. Particular examples are the hydrogen atom in external magnetic fields[9,10], crossed electric and magnetic fields, van der Waals force[12,13], periodic microwave radiation[7] and so on. It is the aim of this article to bring out the salient features of these developments and to point out the future promises.

Quantum signature of chaos

Energy levels and universality

Bounded quantum systems are characterized by discrete eigenvalues and normalizable eigenfunctions. The macroscopic chaos (namely classical chaos) is essentially due to the long-time behaviour of trajectories in phase space. The Heisenberg's uncertainty relation, namely $\Delta E \cdot \Delta T \geq \hbar$ implies that long-time behaviour should necessarily be associated with short energy intervals. Thus, one possible approach to understand the implications of chaos in quantum systems is to look for short-range correlations between energy levels (spacings). Indeed, by examining a large class of quantal systems[1-10] such as billiards of various types[15], coupled anharmonic oscillators[12,13], atomic and molecular systems[14], it has been realized that there exists generically a universality in the spacing distribution[16] of the quantum version of classically integrable as well as chaotic systems. For regular systems nearest-neighbour spacings follow a Poisson distribution, while chaotic systems follow either one of the three universality classes (depending upon the underlying symmetry and the value of the angular momentum (spin)) given in Table 1. For chaotic systems the three universality classes correspond to Gaussian Orthogonal Ensemble (GOE) or Wigner statistics, Gaussian Unitary Ensemble (GUE) statistics and Gaussian Symplectic Ensemble (GSE) statistics, similar to the ones which occur in random matrix theory[17-19] of nuclear physics developed by Wigner, Dyson, Mehta and others during the sixties. Finally, the near integrable and intermediate cases are found to satisfy either the Brody or Berry–Robnik or Izrailev distribution[20,21].

Energy level dynamics

In fact, one can rigorously analyse the scheme of transition from Poisson statistics to the Gaussian random matrix ensemble statistics (corresponding to transition from regular to chaotic motion in classical dynamics) in the framework of level dynamics[22-24]. Considering a generic Hamiltonian $H = H_0 + \lambda V$, where λ is typically the nonintegrability parameter which is now taken as a 'time' variable, one can deduce the 'equations of motion' for the nondegenerate discrete energy levels and eigenfunctions under the λ-flow from the Schrödinger eigenvalue problem[24] $H\,|\,n(\lambda)\rangle = x_n(\lambda)\,|\,n(\lambda)\rangle$:

$$\frac{\mathrm{d}x_n}{\mathrm{d}t} = \langle n\,|\,V\,|\,n\rangle = p_n \quad (t = \lambda), \tag{1a}$$

$$\frac{\mathrm{d}p_n}{\mathrm{d}t} = 2\sum_{m \neq n} V_{nm} \cdot V_{mn}(x_n - x_m)^{-1}, \tag{1b}$$

$$\frac{\mathrm{d}\,|\,n\rangle}{\mathrm{d}t} = \sum_{m \neq n} |\,m\rangle\, V_{mn}(x_n - x_m)^{-1}, \tag{1c}$$

Table 1. Various universality classes of level statistics

Classical dynamics	Symmetry	Quantum level statistics	Examples
Regular	–	Poisson	Integrable billiards, (un)coupled oscillators, etc.
Near-integrable	–	Intermediate (Izrailev, Berry–Robnik, Brody)	Near-integrable cases of coupled oscillators
Chaotic	Time reversal invariance	GOE (Wigner)	Hydrogen atom in strong magnetic field, hyperbolic systems, anisotropic Kepler system
	Broken time reversal invariance	GUE	Aharanov-Bohm billiard
	Time reversal invariance + (1/2) odd integer total angular momentum	GSE	Neutrino billiard

$$\frac{d\langle n|}{dt} = \sum_{m \neq n} \langle m|\, V_{nm}(x_n - x_m)^{-1},$$

$$V_{nm} = \langle n | V | m \rangle. \tag{1d}$$

Equation (1) can be shown to be equivalent[24] to a completely integrable N-particle Calogero–Moser system endowed with an additional internal complex vector space structure. The solutions of this system determine the possible energy spectrum and wavefunction patterns at an arbitrary flow value of λ, the nonintegrability parameter. Further, one can use these solutions to describe the origin of successive avoided crossings observed in the energy level structure of bounded systems[2,25]. Then by making use of a grand canonical ensemble one can derive the universal spacing distributions as in the case of random matrix theory[25–27]

Wavefunction behaviour: Scarring and localization

The phase trajectories of classically integrable and near integrable Hamiltonian systems can be easily seen to lie on energy shells corresponding to invariant tori, the so-called KAM tori[28]. Correspondingly, in the semiclassical limit, the Einstein–Brillouin–Keller (EBK) quantization rules hold good and the quantal eigenstates are known to be localized[28] on the invariant tori. When the classical system is chaotic, the phase trajectories traverse the accessible phase space ergodically. Typically, the chaotic trajectories consist of infinite number of unstable periodic orbits (the stable periodic orbits have zero measure). Surprisingly, the corresponding quantum eigenfunctions in generic examples have also been found to have structures with high amplitude along short, unstable periodic orbits. Heller[29,30] described them as having 'scars' of classical periodic orbits and provided theoretical arguments for the constructive quantum interference responsible for their appearance. The enhanced probability density of the scarred wavefunctions is ascribed to the focal points of the periodic orbit. The scars play an important role in interpreting experimental results[31], for example modulation of intensity.

A second, but related, aspect of quantal wavefunctions of classically chaotic system is 'localization'. By considering a periodically kicked rotator, it has been shown by Casati and Molinari[32] that while in the classical case the energy diffuses as a function of time, in the quantal case, after a finite time, the energy (diffusion) displays a bounded fluctuation about a finite mean value. By analogy with Anderson localization in solid-state physics, one can argue that the quantum-mechanical suppression of diffusion is essentially due to the localization of the wavefunction[33] arising from the interference effects.

Rydberg atoms and quantum chaos

Atomic systems are considered to be the most suitable testing grounds[10,14] for understanding the various ideas on quantum chaos discussed above. One of the reasons behind this is that the underlying systems are amenable to classical, semiclassical and quantum treatment. They are also experimentally realizable in the laboratory. Moreover, one can make a systematic experimental and theoretical (analytical and numerical) comparison.

We know that hydrogen atom is the simplest atomic system containing only one electron. Its classical analogue is the Kepler problem of a planet moving around a relatively stationary star under the action of the gravitational force. Both the quantum and classical problems are solvable and comparisons can be made. In the presence of a physically realizable and meaningful external field, the classical analogue of the hydrogen atom (the perturbed Kepler problem) shows a rich variety of nonlinear phenomena[9,34] and is a typical system exhibiting Hamiltonian chaos[34,35]. The corresponding quantum system represents the so-called highly excited Rydberg atoms (effective one-electron atoms, where the outermost electron is excited to a higher energy level so that the remaining electrons with the nuclear core form an effective nuclear charge) in external fields. Thus, the quantum manifestations of classical chaos can be inferred from the study of such real atomic systems in external fields.

Quadratic Zeeman effect

To appreciate that the Rydberg atoms in external fields are indeed laboratory-realizable systems, let us consider for a moment the problem of hydrogen atom kept in a constant magnetic field along the z-direction. The associated Hamiltonian can be written as

$$H = \left(\frac{(p - eA/c)^2}{2m}\right) - \left(\frac{e^2}{r}\right), \tag{2}$$

where the field is derived from the vector potential $\boldsymbol{A}$ as $\boldsymbol{B} = (\nabla \times \boldsymbol{A}) = (0, 0, B)$, $\boldsymbol{A} = \frac{1}{2}(\boldsymbol{B} \times \boldsymbol{r})$. Here m is the reduced mass, $\boldsymbol{p}$ the momentum and e the electronic charge. Then one can rewrite the Hamiltonian (2) as

$$H = \frac{p^2}{2} - \left(\frac{e^2}{r}\right) + \omega L_z + \tfrac{1}{2} m\omega^2 (x^2 + y^2), \tag{3}$$

where L_z is the z-component of the angular momentum $(= xp_y - yp_x)$ and ω half the cyclotron frequency,

$$\omega = \tfrac{1}{2}\omega_c = \frac{eB}{2mc} \tag{4}$$

Thus, when the magnetic field strength is small, the last term on the right-hand side of equation (3), the so-called quadratic Zeeman term, can be neglected and we have the well-known anomalous Zeeman effect[36]. In order for this quadratic term to have an appreciable effect, it should be of the order of the Coulomb interaction. An estimate of the required field strength can be obtained as follows.

Comparing the Rydberg energy (i.e. the ground-state energy $R = (-me^4/2\hbar^2)$) with the (two-dimensional) oscillator energy ($\hbar^2\omega$) gives a critical field strength of

$$B = B_0 = \frac{m^2e^3c}{\hbar^3} \approx 2.35\times 10^9\,\mathrm{G} = 2.35\times 10^5\,\mathrm{T}. \quad (5)$$

Thus, one requires an astronomical field strength of 2.35×10^5 T in order to realize the effect of the quadratic Zeeman term, which in the classical case leads to chaos. However, one can quickly realize that with the recent advent of tunable lasers, atoms can be excited to higher levels, even up to the order of the principle quantum number $n \approx 50$ to 100, so that there is a dramatic reduction of the field strength required in the laboratory, as given in Table 2. It is clear that for $n = 50$ state, just a 2 T field, comparable to that of the Coulomb field, is enough to realize the effect of chaos.

In fact, a large body of investigations has been performed both analytically[8–10, 37] (theoretical and numerical) and experimentally[38] to realize the quantum effects of chaos in the quadratic Zeeman problem. We summarize the details below.

The motion of the electron is regular in a weak magnetic field and chaotic on a large scale in a strong magnetic field. In the intermediate region there is a smooth transition. As far as the energy spectrum of the corresponding quantum system is concerned, in the weak-field regime the nearest-neighbour spacing distribution follows Poisson distribution, whereas at the strong field regime they follow the GOE statistics. In the intermediate regime, where both regular and chaotic trajectories coexist, it is described by intermediate statistics.

In the chaotic region near the ionization limit Garton

Table 2. Variation of certain physical entities of Rydberg states as a function of the principle quantum number n[10]

Physical entity	Scaling relation	$n = 1$	$n = 50$
Size	n^2	1 Å	0.5 μm
Energy	$1/n^2$	13.6 eV	5 meV
Electron velocity	$1/n$	2×10^6 m/s	4×10^4 m/s
Critical electric field	$1/n^4$	10^9 V/cm	100 V/cm
Critical magnetic field	$1/n^3$	2×10^5 T	2 T

and Tomkins[39] observed a series of broad unresolved resonances called quasi Landau resonances equally spaced by 1.5 cyclotron frequency. This phenomenon shows that the long-range correlations exist in the spectrum which can be analysed using the so-called Gutzwiller's trace formula, which involves the summation over all classical periodic orbits[1]. The experimentally observed spectra[38] are shown to have an impressive agreement with these classical periodic orbits.

By making use of Husimi distribution in phase space (which represents the overlap of the eigenstate with a coherent state), the quantum localization near the classical invariant torus is identified[10] for weak-field limit whereas in the strong-field limit no such localization is observed. Thus, there is a qualitative difference between regular and chaotic eigenstates of the quantum systems.

Hydrogen atom in other external fields/interactions

It is not only the problem of hydrogen atom in a constant magnetic field that is of interest in quantum chaos. There are a host of other interesting problems in which the hydrogen atom interacts with external fields which are also of considerable interest both from theoretical and experimental points of view. They are enumerated in Table 3.

Hydrogen atom in a generalized van der Waals interaction

If we consider two identical atoms (neutral) separated by a distance d (which is large in comparison with the radius of the atoms), then the atoms induce dipole moments on each other, which will cause an attractive interaction between them. This is the so-called van der Waals interaction or London interaction or induced dipole–dipole interaction. It is possible in this case to show that the interaction between the hydrogen atom and a nearby metal surface (called the instantaneous van der Waals interaction) can be described[11, 41] by the Hamiltonian

$$H = \frac{p^2}{2} - \left(\frac{1}{r}\right) + \gamma_l\,(x^2+y^2+2z^2), \qquad \gamma_l = -1/16\,d^3. \quad (6)$$

By generalizing the above and equation (3), Alhassid *et al.*[42] have introduced the so-called generalized van der Waals interaction with the Hamiltonian

$$H = \frac{p^2}{2} - \left(\frac{1}{r}\right) + \gamma(x^2+y^2+\beta^2z^2), \quad (7)$$

which encompasses many of the physically interesting

Table 3. Various physically meaningful perturbations of hydrogen atom. $H = H_0 + H_1$; $H_0 = (p^2/2) - (1/r)$

Physical problem	Typical form of H_1	Properties	Ref.
Stark effect	Fz	Separable, no chaos	37
Magnetic field (constant)	$\omega L_z + (1/2)\, m\omega^2 (x^2 + y^2)$	Nonintegrable; order to large chaos smooth transition	10
Oscillating electric field	$Fz \,.\, \cos(\omega t)$	Order to chaos depending on F and ω values (nonintegrable)	7
Crossed electric and magnetic field (constant)	$\gamma (x^2 + y^2) + Fz$	Depending on γ and F value either regular or chaotic (nonintegrable)	40
Spherical quadratic Zeeman problem	$\gamma (x^2 + y^2 + z^2)$	Separable, no chaos (integrable)	34
Instantaneous van der Waals field	$\gamma (x^2 + y^2 + 2z^2)$	Near-integrable order to small-scale chaos transition	12
Waveguide	$\gamma (x^2 + y^2 + \alpha z^2)$, $\alpha = [2, 8/3]$	Near-integrable, order to small-scale chaos transition	41

F, γ, ω and α are parameters.

systems discussed above for appropriate choices and range of the parameters[13] γ and β. Interestingly, this system has also a close analogy with the Paul trap problem of ions in precision atomic spectroscopy[43]. In a Paul trap, an rf electric field is applied between the end plates and a ring-shaped electrode at the centre so that the fields create hyperbolic potentials in which the motion of ions is harmonic to a first order. At the centre of the trap, the ion inhabits virtually a field-free region. The Hamiltonian of such a Paul trap[44] is essentially given by

$$H = \frac{p^2}{2} + \frac{1}{r} + \frac{1}{2}\,(x^2 + y^2 + \beta^2 z^2)\,, \tag{8}$$

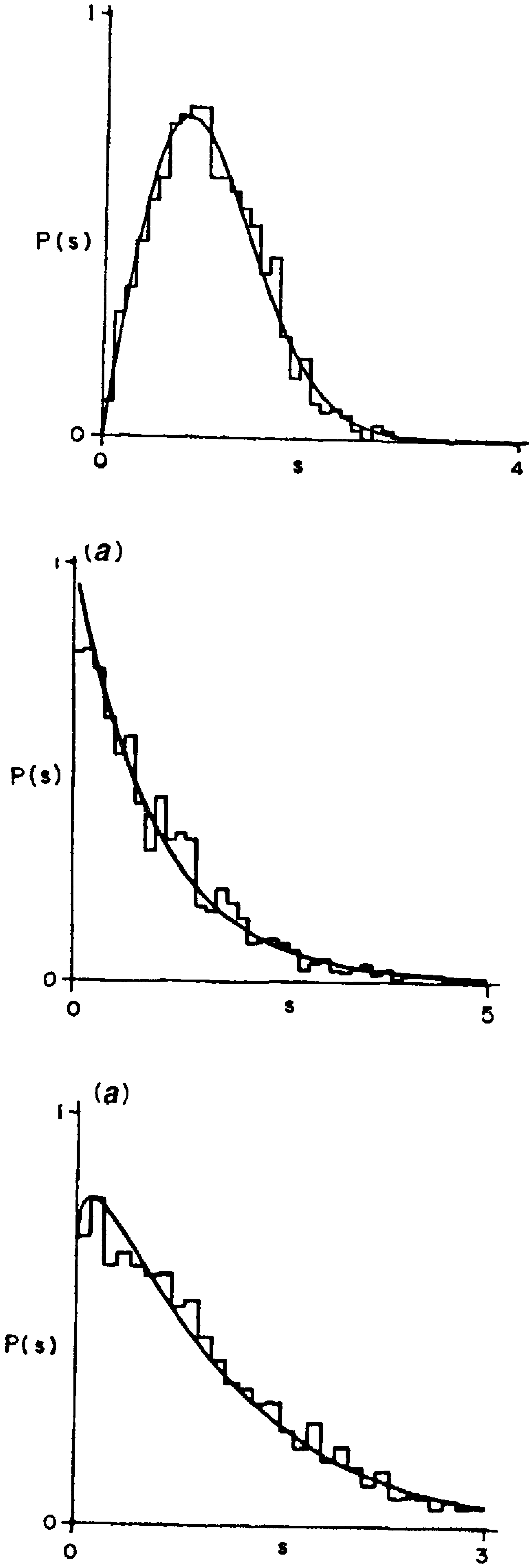
where β represents the deformation of the secular oscillator. One may note that equation (8) is analogous to equation (7) except for a change in sign of the Coulombic term.

Hamiltonian (7) is not an exactly solvable problem for arbitrary values of β. By using cylindrical and semiparabolic coordinates one can show that the z-component of the angular momentum is a constant of the motion so that the system becomes effectively a problem of two coupled sixth-power anharmonic oscillators[45]. Then all the recently developed techniques to deal with chaotic, nonintegrable Hamiltonian systems can be used to study the classical and semiclassical aspects. Detailed analysis[34] shows that as the parameter β increases for arbitrary γ, there is a chaos–order–chaos–order–chaos–order–chaos type of transition, with exactly integrable behaviour at $\beta = 1/2$, 1 and 2 due to the existence of certain nontrivial dynamical symmetries of the system. The corresponding quantum problem can be analysed by solving the Schrodinger equation numerically in terms of the SO(4, 2) group generators using a scaled set of normalized Coulombic wavefunctions[43]. In this way a large number of converged eigenvalues can be obtained and the level statistics can be analysed. Again as the parameter β increases, one obtains a GOE Poisson–Brody–Poisson–Brody–Poisson–GOE type of transition corresponding to classical dynamics, thereby confirming the classical and quantum connections[43]. Some representative details are given in Figures 1–3. Similar studies can also be performed for wavefunction dynamics.

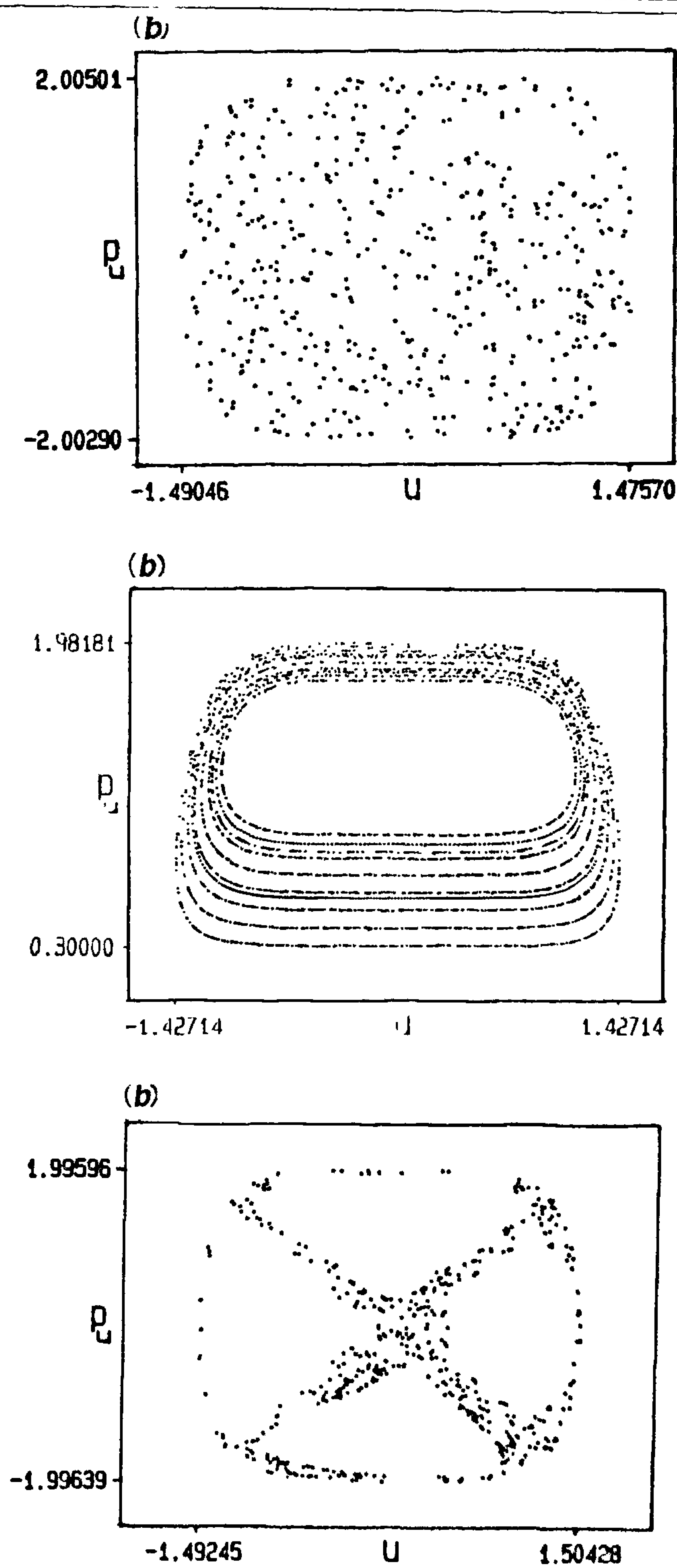
Outlook

It is not only the study of Rydberg atoms that is of interest in quantum chaos. Recent investigations show that investigations of Rydberg molecules are also of paramount importance in view of the nonuniversality properties of the correlations between intensities and spacings[46] and the classical chaotic autoionization mechanism proposed for the experimentally observed ionization process[47]. In a related development, for the He atom for collinear configuration with both electrons on the same side of the nucleus, one finds regular behaviour, while for electrons on different sides of the nucleus, fully developed chaotic behaviour occurs, whereas the motion on the Wannier ridge shows a mixed behaviour. Interestingly, all these three regions have been quantized semiclassically for the first time. Moreover, there seems to exist a close agreement with pure quantum-mechanical and experimental results[48]. Also, at present many interesting classical, semiclassical and quantal investigations are ongoing along this direction.

In the case of mesoscopic systems (whose size $\approx 10^{-9}$ m), by looking at the dispersion of energy levels to external perturbations (like magnetic field), the so-called generalized conductance was recently introduced. Indeed remarkably, the conductivity properties



Figures 1 *a*–3 *a*



Figures 1 *b*–3 *b*

Figures 1 *a*, 2 *a*, 3 *a*. Nearest-neighbour spacing distribution of the hydrogen atom in a generalized van der Waals potential problem for $\beta = 1/4$, $1/2$ and $\sqrt{0.4}$; **Figures 1 *b*, 2 *b*, 3 *b*.** Poincaré surface of section of Hamiltonian (7) (using the corresponding oscillator counterpart Hamiltonian) for $\beta = 1/4$, $1/2$ and $\sqrt{0.4}$.

of these mesoscopic systems share analogous universality properties exhibited by quantum chaotic systems[49]. These findings have brought much practical importance in the field of 'quantum chaos'.

Finally, many interesting results are coming up in the quantum effects of chaotic scattering processes. For example, the so-called Ericson fluctuations in scattering cross-sections are related to the classically chaotic trajectories and also they show universal behaviour. The observed resistance measurements of microstructures like quantum dots, quantum wires, etc., cannot be explained by standard solid-state physics textbook style but one can explain them using simple quantum scattering aspects[50]. It appears that quantum manifestations of classical chaos will indeed have important practical applications.

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