Highly excited atoms reveal the limitations that quantum interference imposes on the dynamics of classically chaotic systems

Quantum mechanical suppression of chaos

The relation between determinism and predictability is the central issue in the study of 'deterministic chaos'. Much knowledge has been accumulated in the past 10 years about the chaotic dynamics of macroscopic (classical) systems. Our intention is to examine the implications of chaos in the microscopic quantum world -- in other words, how to reconcile the correspondence principle with the inherent uncertainties which reflect the wave nature of quantum dynamics. Some rather surprising and counterintuitive results emerge from this effort which bear an intriguing similarity to a phenomenon associated with electric conductivity in disordered solids, namely 'Anderson localisation'. It is very rewarding that these developments do not remain in the lofty realms of abstract theoretical physics. Indeed, recent atomic physics experiments demonstrate clearly that chaos is relevant to the microscopic world. In particular, such experiments emphasise the urgent need to clarify the genuine quantum mechanism which imposes severe limitations on quantum dynamics, and renders it so very different from its classical counterpart.

The first question to be addressed is the rather confusing concept of 'deterministic chaos'. This seems to be a contradiction in terms since chaos is usually considered as an antonym of determinism. The following example should hopefully convince the reader that this is, in fact, an appropriate name.

A schematic experiment which we shall analyse in some detail throughout this article is shown in figure 1. It is very similar to an actual experimental set-up used by Meissner and Schmidt in 1986 for the study of the onset of classical chaos. A magnetic needle is free to turn about a vertical and frictionless axis. When a periodic train of current pulses is sent through the coils, the needle is exposed to a periodic succession of magnetic field bursts. Each burst lasts only a very short time and the peak field intensity can be adjusted to any desired value. In the time interval between successive bursts the field vanishes. The magnetic needle is at rest before the field is applied, and its orientation is $\theta_0$. After $N$ field bursts the amount of kinetic energy $E(N)$ acquired by the needle is measured, although it is sometimes advantageous to consider the mean energy $\langle E(N) \rangle$ obtained by averaging over all possible initial orientations of the needle.

In spite of the completely deterministic nature of this system, the outcome of the experiment is best described in statistical terms. Moreover, the long-time behaviour of the system cannot be described by any other means. To substantiate this claim, let us first analyse our model by applying Newton's laws of mechanics. Suppose that just before the $n$th field burst the needle was at an orientation $\theta_n$ and it was turning about its axis with angular momentum $I_n$. During the short duration of the $n$th field burst the orientation of the needle hardly changes at all. The applied torque adds $k \sin \theta_n$ to the angular momentum, where $k$ is the product of the dipole moment and the time integral of the field during a burst. During the quiescence period between field bursts, the angular momentum is not altered, and its value determines the change in the orientation due to the free turning of the needle. The orientation angle changes by the amount $I_{n+1}/J$ where $J$ is the moment of inertia of the needle.

With all this information, we can relate the values of the angular momentum and the angle just before the $(n+1)$th field burst to their values just before the $n$th burst by the equations

$$ I_{n+1} = I_n + k \sin \theta_n $$

$$ \theta_{n+1} = \theta_n + I_{n+1}/J $$

(1)

These relations determine completely and unambiguously the motion of the needle. They can be solved iteratively, and give the angular momentum and orientation ($I_N$, $\theta_N$) after any number of bursts $N$ once the initial conditions ($I_0$, $\theta_0$) are specified. This is a very simple iteration procedure which can even be solved on a pocket calculator or a simple home computer. It is a recommended bait to lure the innocent into the study of chaotic dynamics since it displays many of the elegant and surprising features of chaos despite its structural simplicity. Many systems can be described in terms of iteration schemes of this kind, with the picturesque generic name 'kicked rotors'.

The 'random' character of the needle dynamics becomes apparent when one observes the dependence of the angular momentum on time. Some typical histories (which differ
only in the initial orientations $\theta_i$ are shown in figure 2. Can you notice any order in these functions? Could you believe that they are the results of a deterministic iterative scheme? Now that we see that the term ‘deterministic chaos’ is indeed called for, let us try to understand the mechanism which induces it.

 Angular sensitivities

Two ingredients are necessary and sufficient to move any respectable system into the chaotic domain. The first is the extreme sensitivity to small changes in the initial conditions. The red and the blue histories in figure 2 are classical trajectories which differ by less than one thousandth of a degree in the initial value $\theta_i$. Yet, after approximately 15 field bursts they cannot be considered similar any more. In other words, any small difference between initial conditions is amplified very rapidly, so that any memory of previous ‘neighbourhood’ fades away after a few field bursts. This exponential amplification of small deviations is termed ‘stretching’. The second mechanism shows itself in the needle dynamics in the following way. To know how the needle is orientated just before the next burst, one does not need to count how many revolutions it completed between the bursts. What matters therefore is the value of the angle modulo 360°. The modulo operation (or ‘folding’) forces us to concentrate our attention on the less significant digits in our numbers. The leading digits are wiped out by the folding operation.

Any random number generator on your computer operates on the basis of stretching and folding. This is how this completely deterministic instrument produces strings of ‘random’ numbers. The motion of the needle, when the magnetic field is sufficiently strong, is also subjected to stretching and folding. Hence it has as much of a claim to the title ‘chaotic’ as your random number generator has to the title ‘random’.

We can now return to the original problem with which we started and calculate the energy gained by the needle from the action of the driving force. This we do by iterating equation 1 for any desired number of bursts, setting $I_e = 0$ and averaging over the initial orientations $\theta_i$. The results are shown in figure 3. The ensemble of initial conditions consists of 100 values for $\theta_i$ and for each we show the value of the angular momentum after 5 bursts (red), 25 bursts (green) and 125 bursts (black). One can clearly see that the initial cloud of points on the $I = 0$ axis diffuses in time and becomes broader as the number of bursts increases. The kinetic energy after $N$ bursts is given by $E(N) = I_e^2/2J$ and its mean value can be calculated easily (see figure 4). The mean energy increases linearly with $N$. This linear dependence is a typical feature of diffusion which is usually observed in problems involving random walks. It is an important signature of the underlying chaotic dynamics.

The rate of diffusive energy transfer is termed the diffusion constant $D_i$ in the needle problem and for sufficiently strong fields $D = kT/4J$.

The kicked rotor is a paradigm of a large class of periodically driven systems, which become chaotic when the external driving force exceeds a critical strength. In all these systems, one of the most conspicuous manifestations of chaos is the diffusive energy transfer. Of most practical importance are the systems of atoms or molecules which are exposed to strong electromagnetic fields. By analogy with the above one could infer that for field strengths which induce chaotic dynamics, an arbitrarily large excitation energy can be
4 Dependence of the mean energy of the ensemble shown in 3 on the number of bursts. The fluctuations about the straight line are due to the finite number of points in the sample.

The fly in the ointment is that atoms and molecules are governed by quantum mechanics and the quantum solution of the present problem has a surprise in store.

Quantum interference

Quantum theory prohibits unlimited diffusive energy transfer; the amount of energy that can be transferred is bounded. A computer simulation (published by Casati and colleagues in 1979) shows that during the initial exposure time quantum and classical theories match quite well (figure 5). However, after some time this agreement breaks down, and the quantum energy displays bounded fluctuations about a finite mean value.

The suppression of classical diffusion by genuine quantum effects is already known in quite a different physical context. Think of electrons in a disordered solid: at low temperatures the electrons scatter elastically from the randomly placed atoms. They perform a random walk, and classical mechanics predicts that an initially localised electron cloud would diffuse over the entire sample after sufficiently long time. Anderson was the first to realise that quantum interferences may suppress the diffusion to such an extent that the electrons are 'localised' and the material is an insulator, whereas it would have been a conductor if Newtonian mechanics were sufficient.

Other important features which characterise the quantum localisation phenomenon are relevant in the present context: the final distribution of the electron cloud is not smooth as a function of the distance from its centre of gravity. Instead, strong density fluctuations appear intermittently due to constructive and destructive interferences.

The effectiveness of localisation depends on the dimension of the space in which the electron is allowed to move. If it is confined to move on a line, localisation will be very effective and it will always overcome classical diffusion. In three dimensions, however, a transition between localised and non-localised quantum behaviour is expected.

One can 'delocalise' the electron and restore conductivity by introducing noise (inelastic collisions, interaction with thermal phonons or whatever). Sensitivity to external noise is expected since localisation is due to an intricate interference phenomenon which is destroyed once all phase relations are blurred by the noise. (A clear distinction needs to be made here between 'disorder' and 'noise'. Disorder only means that in a given sample one cannot specify the positions and compositions of the scatterers by a simple rule. Noise interferes with the dynamics by exposing it to external random forces. In the present context, disorder is 'good' since it induces quantum localisation, whereas noise is 'bad' since it destroys it.)

It should be emphasised that Anderson localisation appears whenever waves scatter from randomly distributed scatterers, and it is not only expected to occur in quantum systems. It is also encountered in, for example, scattering of sound and water surface waves, as well as electromagnetic waves off randomly placed scatterers. The motion of the quantum electron in a disordered solid is just a particular example of this very general wave phenomenon.

At first sight it seems hard to understand why Anderson localisation is relevant at all to our quantised kicked rotor. In particular, it is not clear which element in the latter system is responsible for the disorder which is so crucial in inducing Anderson localisation. In 1982 Fishman, Grempel and Prange were the first to realise the connection. They showed that a simple version of the Anderson theory for spatial localisation in one dimension can be carried over to describe localisation on the angular momentum axis of a certain kicked rotor. They also proposed that the disorder is due to a mechanism reminiscent of the 'folding' operation which is one of the necessary conditions of classical deterministic chaos. This mechanism imparts 'quasi-random' phases to the amplitudes which interfere and produce localisation. One of the most important open problems in the field is to understand precisely how this happens, and which are the ingredients of the underlying chaotic dynamics that are necessary to induce localisation.

As long as a complete theory is absent, one has to rely mostly on computer simulations of the quantum dynamics. These provide very substantial numerical evidence in favour of the applicability of Anderson's theory of localisation to the description of the kicked rotor. In particular:

- The diffusive energy transfer is suppressed.

5 A comparison between the classical (---) and quantum mechanical (-----) evaluation of the mean energy of the needle as a function of time. (After T Dittrich and R Graham 1989 Naturwissenschaften 76 401)
approximated by a Rydberg series. Classically, the highly excited electron can be considered as moving on a classical Kepler orbit with micron dimensions. Besides testing localisation theories, the study of Rydberg atoms, therefore, is an interesting subject in itself.

In the localisation experiment the atoms traverse a cavity where they are exposed to a microwave field to provide the periodic driving. In the interaction zone, the atoms are typically exposed to a few hundred microwave field cycles. During this time the atoms absorb energy from the field: the Rydberg electron remains bound to the nucleus as long as the gained energy does not exceed the binding energy. If it does the atom ionises and ions emerge from the cavity. One can therefore use the ionisation probability (i.e. the fraction of ions emerging from the cavity) to test the process of energy gain from the field. Classical mechanics predicts that ionisation will occur with a high probability once the field strength is higher than a threshold value which marks the onset of chaotic dynamics which, in turn, induces a diffusive energy gain. Quantum localisation would be observed if the field strength required to induce ionisation exceeds the classical threshold (Casati and colleagues 1986).

The pioneering experiment of Bayfield and Koch at Yale in 1976 which drew attention to this particular system was recently repeated and greatly improved by Koch’s group at Stony Brook and by Bayfield and colleagues at Pittsburgh. Threshold fields were measured for Rydberg atoms which were prepared in initial states in the range $60 < n < 90$. The results clearly show the effect of Anderson localisation (figure 6) since the observed ionisation thresholds exceed classical predictions. The results in figure 6 come from a collaboration between the experimentalists in the Pittsburgh group and theoretical analysis by the Milano-Novosibirsk team of Casati, Guarneri and Shepelyansky.

Other experiments by Koch and colleagues in 1986 showed that the ionisation probabilities did not grow monotonically with the field strength. Instead, at field values near and below the classical ionisation threshold, enhanced ionisation was observed. These enhancements are due to the same mechanism which brings about the strong fluctuations in Anderson localisation.

Ionisation experiments, however, give only an indirect hint to the existence of Anderson localisation. More direct evidence was provided recently by the measurement of the distribution of excitation probabilities of the high-$n$ states for different exposure times. In these experiments (conducted by L Sirko and H Walther and their group in Munich) it was observed that, after a certain transient time, the ionisation probability distribution does not change with increasing interaction time despite the fact that strong driving fields were applied! What happens is that destructive interference prohibits further transitions to highly excited states and, as a result, the atom is forced to stay in relatively low excited states independent of the time it spends in the external fields – a clearcut demonstration of Anderson localisation.

In the same experiments, the effect of noise on localisation was measured by admixing electronic noise to the output of the highly stable microwave generator. (Such an experiment is very difficult to carry out, not because of technical problems but because one has to convince the

---

**Experimental ionisation thresholds measured for various high initial quantum numbers $n_0$. The abscissa is the field frequency scaled by the classical Kepler frequency $1/n_0^2$. Threshold fields are given in units of the mean atomic field experienced by the electron in the Kepler orbit ($\sim 1/n_0^2$). The symbols show experimental threshold values measured at different scaled field frequencies. The dotted line is the predicted classical theory. The dashes are the result of quantal calculations, whereas the solid line corresponds to an approximate localisation theory applied to the present system. (J E Bayfield et al 1989 Phys. Rev. Lett. 63 364)**

- The distribution of excitation energies displays large fluctuations which are very sensitive to small variations in the parameters of the driving force.
- Studies of systems with more degrees of freedom (e.g. a few more needles are added which interact among themselves through the dipole–dipole interaction) reproduce the expected dependence of the localisation properties on the dimensionality.
- The localisation phenomenon is destroyed if some noise is allowed to affect the system, simultaneously with the periodic driving force.

**Atomic tests**

As soon as these surprising theoretical results were discovered, it became clear that the experimental and practical consequences of quantum mechanical localisation in classically chaotic systems should be demonstrated.

The closest analogue of the needle system, taken from molecular physics, was proposed by S Fishman and us in 1986. Polar diatomic molecules (such as CsI with its large electric dipole moment and moment of inertia) act as the rotor. The kicking is supplied by a train of periodic electric field bursts generated by a superposition of the first seven harmonics of a 5 GHz microwave field in a cavity. The experimental set-up should consist of a cooled molecular beam (rotation temperature $\sim 1$–2 K) which undergoes excitation in the microwave cavity and is subsequently scanned by selective fluorescence to yield the distribution of angular momenta in the emerging beam. Detailed calculations have shown that the molecular analogue of the kicked rotor grasps all the important features of the schematic needle system. Although technically feasible and quite important as a possible molecular physics manifestation of Anderson localisation, this experiment has not yet been carried out, mainly because the required microwave equipment cannot be obtained off the shelf.

The concepts and ideas above were put to experimental test in an alternative system: highly excited atoms periodically perturbed by an external electromagnetic field. The test was performed using hydrogen or alkali atoms whose outer electron was excited typically to principal quantum numbers $n$ ranging from 32 to 90. Highly excited atoms of this type are known as Rydberg atoms, since the excitation energies of the outer electron can be well
experimentalists to reduce the quality of their electronics, which is very much against their professional pride and principles. . . . Once all the problems were overcome, the ratio between the noise and the coherent field intensity could be controlled and the effectiveness by which the external noise destroys the localisation studied. The results of the experiment prove to be consistent with the theoretical predictions.

Universal

The experimental evidence supports the theoretical predictions of the role played by Anderson localisation in the quantum dynamics of classically chaotic systems of the type discussed here. It also provides another important lesson: classical chaos and induced diffusion is a universal phenomenon. It appears in all systems which satisfy a certain minimal number of conditions. In the same way, the suppression of classical chaos by quantum interference appears to be universal. It has been shown above to occur in two very different systems - the hydrogen atom and the magnetic needle. Both systems, however, share one important feature: they are classically chaotic.

The large discrepancy between the quantum and the classical descriptions of the needle dynamics is not at variance with Bohr's correspondence principle. The correct statement of Bohr's principle is that for any given time interval, $\tau$, the quantum and classical theories yield consistent descriptions within the time interval $t < \tau$, if the relevant quantum numbers are sufficiently large. Thus, the domain of high quantum numbers depends on $\tau$. The longer the prescribed time interval, the larger must be the quantum numbers if correspondence is to be satisfied. The discrepancy which we observe in figure 5, for example, is due to the fact that we are looking at the same domain of quantum numbers whilst increasing the time interval. The initial agreement between the quantum and classical results for $t < \tau = 20$ units confirms the correspondence principle.

This research belongs to the newly emerging field of physics termed "quantum chaos" by Michael Berry in 1987. It deals with the quantum mechanical description of classically chaotic systems, and develops at the borderline between quantum and classical mechanics. At present the interesting problems that must be faced outnumber the available paved routes and standard tricks, but the questions that are asked are relevant to fields ranging from nuclear to molecular and solid state physics.

Further reading

R Blämel and U Smilansky 1987 Z. Phys. D 6 83
B Chirikov 1979 Phys. Rep. 52 263

Reinhold Blämel is in the chemistry department, University of Pennsylvania, Philadelphia, PA 19104-6323, USA and Uzy Smilansky is in the nuclear physics department, Weizmann Institute, 76100 Rehovot, Israel