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Resonance scattering in a two-dimensional non-integrable system

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Abstract. Resonance eigenvalues of a two-dimensional non-integrable system whose Hamiltonian can be expressed in terms of a polynomial of the position and momentum operators are evaluated by a method based on complex scaling. To find the eigenvalues of the complex-scaled Hamiltonian, we modified the Householder and the QR methods to treat a complex symmetric matrix. Resonance eigenstates over the potential barriers are primarily considered. We show how the non-linear effects, e.g. the chaotic behaviour in classical mechanics reflect the quantum system by investigating the distribution of the resonance eigenvalues on the complex energy plane.

1. Introduction

This paper is concerned with the quantum mechanical motion of a particle in a two-dimensional 'open' system whose energy surface is unbounded in phase space. In contrast to a closed Hamiltonian system in which all classical trajectories are bounded, most of trajectories of an open system eventually leave the scattering region. Even in the case of an open system, however, if we can separate its Hamiltonian into one-dimensional parts, there exist periodic orbits with an infinite lifetime. Recently, Bleher *et al* (1990) showed that under special initial conditions a non-integrable system can still have orbits trapped in the scattering region for an arbitrarily long time by repeated reflections on the potential walls. (These orbits are not periodic but chaotic in a time-asymptotic sense.) Such classical orbits with a long lifetime may correspond to resonance states in quantum mechanics. Compared to the resonance tunnelling (i.e. the resonance phenomenon in the classical bounded energy region), there have not been sufficient investigations of the resonance scattering.

Resonances associated with poles in the non-physical Riemann sheets of the Green function play a dominant role in understanding the quantum dynamics of a particle in an unbounded system. The positions $\{E_n\}$ and widths $\{\Gamma_n\}$ of the resonances embedded in the continuous spectrum are associated with the resonance eigenvalues $\{\tilde{E}_n\}$ by the relation $\tilde{E}_n = E_n - i\Gamma_n/2$. The complex coordinate method established by Aguilar and Combes (1971), Balsler and Combes (1971) and Symon (1972, 1973) enables us to evaluate the resonance eigenvalues by a transformation of poles on the higher Riemann sheets into the first Riemann Sheet. This transformation is achieved by a rotation of the cuts of the Green function using complex scaling for the position and momentum operators $\hat{q}' = r\hat{q}\exp(i\theta)$ and $\hat{p}' = r^{-1}\hat{p}\exp(-i\theta)$. (This scaling transforms the original Hamiltonian as an Hermitian operator into a complex

symmetric form.) The complex analogue of the usual variational principle, in which the scaling parameters $\{r, \theta\}$ are also regarded as variational parameters, is available to find the resonance states. Many numerical procedures based on the method have been developed in recent years (Moiseyev *et al* 1978, 1984, Moiseyev and Weinhold 1980, Moiseyev 1982, 1983, Milfeld and Moiseyev 1986, Kolin *et al* 1988, Elander *et al* 1982, Rittby *et al* 1983, and Waite and Miller 1981).

In spite of a number of successful developments of the method for one-dimensional systems, only a few attempts have so far been made for multi-dimensional systems in which many interesting phenomena are expected to occur. (In classical mechanics, most of the interesting phenomena including the chaotic behaviour are observed in systems with more than one-dimension.) Waite and Miller have investigated the resonance states under or near the potential barriers in the Hénon–Heiles system, in connection with the mode specificity in unimolecular reaction dynamics (Waite and Miller 1981). They showed that the quantum rate constants due to the tunnelling do not relate to the mode specificity and are irrelevant to the classical quasiperiodic/ergodic behaviour of the system. In this energy region, any classical particle has an identical (infinite) lifetime regardless of the type of motion (e.g. quasiperiodic, ergodic, etc). So we can assume that the type of motion also hardly affects the quantal lifetime of a particle. On the other hand, a classical particle with energy over the barriers may have a finite lifetime (because it may actually escape from the central part of the system.) This classical lifetime can take a wide range of values, strongly depending on the type of trajectory. In this energy region, we can also expect a variety of quantal lifetimes. We think that at energies over the barriers it is meaningful to compare the classical and the quantal motions of a particle in the non-separable system with the motions in the separable one. The purpose of this paper is to examine the dynamics of a quantum mechanical particle in a multi-dimensional open potential through the investigation of the resonance states and to compare it with the corresponding classical dynamics.

We develop a method applicable to any system whose Hamiltonian is expressed in a polynomial of the position and momentum operators. Such a Hamiltonian can be rewritten in terms of a normal product form of the boson annihilation and creation operators. This transformation enables us to use a basis constructed by the number states, with which any matrix element of the Hamiltonian can be analytically evaluated. In addition, the complex scaling can be included simultaneously in the transformation of the operators. For the diagonalization of a scaled Hamiltonian with complex symmetry, we apply the modified Householder method and the modified QR algorithm. We are mainly concerned with how the dynamical properties associate with the resonance states of a system. So a global distribution of the resonances is more important than the accurate position and width of a special resonance. Thus, we determine the scaling parameter $\{r, \theta\}$ so that the majority of the resonances appear in the first Riemann sheet. The 'trajectory and iteration' method by Moiseyev and co-workers (1978, 1980, 1982, 1983, 1984, 1986, 1988) is not adopted.

The next section is devoted to the formalism and computational details to search the resonances of a multi-dimensional open potential. In section 3, we present the results for the two-dimensional model systems and discuss the dynamics of an integrable system and a non-integrable one comparing with the corresponding classical systems.

2. Model systems and the method

As an example of the multi-dimensional problem, we consider the Hénon–Heiles system whose Hamiltonian is written as

$$H_1(\hat{q}, \hat{p}) = \frac{1}{2}(\hat{p}_1^2 + \hat{p}_2^2) + \frac{1}{2}(\hat{q}_1^2 + \hat{q}_2^2) - \epsilon(\hat{q}_1^2\hat{q}_2 + \hat{q}_2^3/3). \quad (2.1)$$

Since the potential of the system is open in the three directions, the system does not have any true bound state even in the energy region under the barriers. Although the eigenstates under the barriers are regarded as bound states in many investigations (Miller 1987, Noid and Marcus 1975, Davis and Heller 1979, and Hose *et al* 1984), they must be resonance states. The properties of the resonance states distributed under and over the barriers are still not clear. By the existence of the coupling term $-\epsilon\hat{q}_1^2\hat{q}_2$, the Hénon–Heiles system is not integrable and shows the chaotic behaviour in classical mechanics (Hénon and Heiles 1964). To compare with the Hénon–Heiles system, we take another system eliminating the coupling term between the two modes:

$$H_2(\hat{q}, \hat{p}) = \frac{1}{2}(\hat{p}_1^2 + \hat{p}_2^2) + \frac{1}{2}(\hat{q}_1^2 + \hat{q}_2^2) - \epsilon\hat{q}_2^3/3. \quad (2.2)$$

To use the number-state basis, we first introduce the transform of the operators $\{\hat{q}, \hat{p}\}$ into the boson annihilation and creation operators $\{\hat{b}, \hat{b}^\dagger\}$ by the relations

$$\hat{b}_k = \frac{1}{\sqrt{2}}(\hat{q}_k + i\hat{p}_k) \quad (2.3a)$$

$$\hat{b}_k^\dagger = \frac{1}{\sqrt{2}}(\hat{q}_k - i\hat{p}_k). \quad (2.3b)$$

Further, we can extend this transformation including the complex scaling by defining the generalized boson annihilation and creation operators $\{\hat{a}, \hat{a}^\dagger\}$:

$$\hat{a}_k = \frac{1}{\sqrt{2}}(r\hat{q}_k \exp(+i\theta) + ir^{-1}\hat{p}_k \exp(-i\theta)) \quad (2.4a)$$

$$\hat{a}_k^\dagger = \frac{1}{\sqrt{2}}(r\hat{q}_k \exp(-i\theta) - ir^{-1}\hat{p}_k \exp(+i\theta)). \quad (2.4b)$$

There exists an orthogonal set $|n_k\rangle$ on which the number operator $\hat{n}_k = \hat{a}_k^\dagger\hat{a}_k$ satisfies the relation

$$\hat{n}_k|n_k\rangle = n_k|n_k\rangle \quad (n_k = 0, 1, 2, \dots) \quad (2.5)$$

because the commutation relation $[\hat{a}_k, \hat{a}_k^\dagger] = 1$ holds. The number-state basis $\{|n\rangle\}$ for our two-dimensional systems is constructed from $|n_k\rangle$ by the relation

$$|n\rangle = |n_1, n_2\rangle = |n_1\rangle \otimes |n_2\rangle. \quad (2.6)$$

With the relations (2.4a) and (2.4b), the Hamiltonians (2.1) and (2.2) are expressed in the normal products:

$$\begin{aligned} \hat{H}(\hat{q}, \hat{p}) &= \hat{H}(\hat{a}, \hat{a}^\dagger; r, \theta) \\ &= V + \sum_i (V_i\hat{a}_i + V^i\hat{a}_i^\dagger) + \sum_{i,j} (V_{ij}\hat{a}_i\hat{a}_j + V^i{}_j\hat{a}_i^\dagger\hat{a}_j + V^{ij}\hat{a}_i^\dagger\hat{a}_j^\dagger) \\ &\quad + \sum_{i,j,k} (V_{ijk}\hat{a}_i\hat{a}_j\hat{a}_k + V^i{}_{jk}\hat{a}_i^\dagger\hat{a}_j\hat{a}_k + V^{ij}{}_k\hat{a}_i^\dagger\hat{a}_j^\dagger\hat{a}_k \\ &\quad + V^{ijk}\hat{a}_i^\dagger\hat{a}_j^\dagger\hat{a}_k^\dagger) \quad (i, j, k = 1, 2). \end{aligned} \quad (2.7)$$

Evaluation of the coefficients V , V_i , V_i^i , V_{ij} , etc is tedious, but straightforward. For example, we have $V_{12}^1 = -2\epsilon r_1^2 r_2 \exp(2i\theta_1) \exp(i\theta_2)$ for (2.1). For brevity, we omit to write details of all other coefficients for (2.1) and (2.2).

Matrix elements of the complex scaled Hamiltonian with respect to the basis $\{|n\rangle\}$

$$\langle n'_1, n'_2 | \hat{H}(\hat{a}, \hat{a}^\dagger; \mathbf{r}, \boldsymbol{\theta}) | n_1, n_2 \rangle \quad (2.8)$$

can be easily evaluated term by term, according to the algebraic relations

$$\hat{a}_k |n_k\rangle = \sqrt{n_k} |n_k - 1\rangle \quad (2.9a)$$

$$\hat{a}_k^\dagger |n_k\rangle = \sqrt{n_k + 1} |n_k + 1\rangle. \quad (2.9b)$$

For example, we get $\langle n'_1, n'_2 | \hat{a}_1^\dagger \hat{a}_1 \hat{a}_2 | n_1, n_2 \rangle = n_1 \sqrt{n_2} \delta_{n'_1 n_2} \delta_{n'_2 n_2 - 1}$. These matrix elements construct a complex symmetric matrix for a Hamiltonian operator such as (2.1) or (2.2).

For a general complex matrix, the diagonalization via a tridiagonal form cannot be usually applied, because the tridiagonalization is not carried out by the unitary transformation. For this case, the method via a Hessenberg form requiring more computational time and memory capacity is conventionally used. However, we develop a procedure diagonalizing the complex symmetric matrix via a tridiagonal form, following the next two steps.

In the first step, the complex symmetric matrix is tridiagonalized by the modified Householder method. In the Householder method, the stepwise tridiagonalization is made by repeating a reflection transformation. In each repeat, the unit normal vector to the reflection plane is needed. For a Hermitian matrix, we can always obtain this vector with the ordinary positive norm. On the other hand, if we keep the norm positive in a complex matrix, a transformed matrix becomes a Hessenberg form. In the modified Householder method developed in this work, we can continue the tridiagonalization of a complex symmetric matrix, normalizing the vector with the pseudo-scalar norm, even though its normalization factor becomes negative or complex. Thus, the tridiagonalization of a complex symmetric matrix can be executed within the same computational effort as in a Hermitian matrix.

In the next step, we use the modified QR method, in which we employ a similarity transformation to diagonalize a tridiagonal matrix, instead of the unitary transformation conventionally used. The convergence of calculated eigenvalues is greatly accelerated by the origin shift. We especially adopt the cubic shift which is calculated as one of the eigenvalues of the lowest 3×3 principal submatrix and is more effective than the quadratic shift widely used. When we carry out the modified QR method, most of the computational time is consumed for calculation of the (complex) square roots as in the case of the standard QR algorithm. We employ an algorithm avoiding the calculation of the square roots.

3. Results and discussion

In our model systems mentioned in the previous section, the anharmonic parameters control the height of the lowest barriers of the potentials and consequently determine the number of the quasi-bounded states lying under (and over) the barriers. (The

height is given by $1/6\epsilon^2$ and we set $\epsilon = 0.2$.) The value of ϵ is much larger than the widely used one for the Hénon–Heiles system ($\epsilon = \sqrt{0.0125}$). If we set $\epsilon = \sqrt{0.0125}$, most of the resonances under the barrier would become almost the true discrete eigenvalues which have extremely sharp peaks.

To clarify the quantum mechanical feature of the two systems, we now investigate the distribution of resonance eigenvalues in the complex energy plane. We use the basis consisting of the 990 number states which are chosen so as to satisfy the inequality $n_1 + n_2 < 45$. The scaling parameters $\{r, \theta\}$ are selected so that the majority of resonances are graphically separated from other points belonging to the cuts. Thus, we settle the scaling parameters as $r_1 = r_2 = 1$, $\theta_1 = 0.05$ and $\theta_2 = 0.05$ for the Hénon–Heiles system. Since the potential of the uncoupled system is not open in the direction of the q_1 -axis, the complex scaling is only needed for the q_2 -axis. Thus, we have $r_2 = 1$ and $\theta_2 = 0.05$ for the uncoupled system.

In figure 1, we show contours of the potential for the uncoupled system (2.2) and a typical quasi-periodic trajectory in the system. Though the particle has excess energy to pass the barrier, its trajectory is quasi-periodic according to the specialized initial position and momentum. This is a characteristic feature for multi-dimensional systems. It comes from the fact that a particle in a multi-dimensional potential may be trapped by repeated reflections on the walls under the favorable initial condition. In figure 2, we also show the shape and the classical trajectory for the coupled system, the Hénon–Heiles system. The tendency to trap a particle is more weakened than in the uncoupled system. As a result of the chaotic (and ergodic) behaviour the particle is randomly reflected at the boundaries and eventually escapes over the walls.

Since the above-mentioned trajectories may strongly depend on the choice of an initial condition, they do not always show clearly the difference in the classical dynamics between the two system. So we consider the distribution of lifetimes of classical particles as a function of their initial conditions. The lifetime of a classical particle can be defined as a time during which it is trapped in a certain region

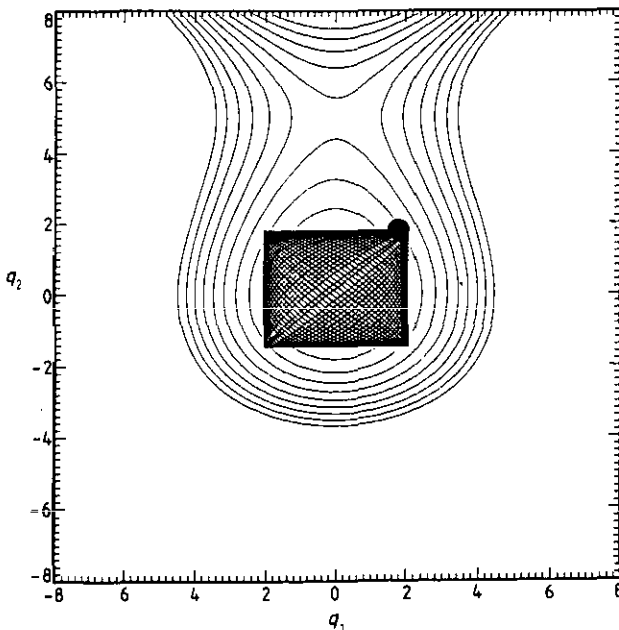


Figure 1. The shape of the potential of $\tilde{H}_2(\hat{q}, \hat{p})$ (which is defined by eliminating the coupling term of the Hénon–Heiles Hamiltonian) together with a typical trajectory in the system.

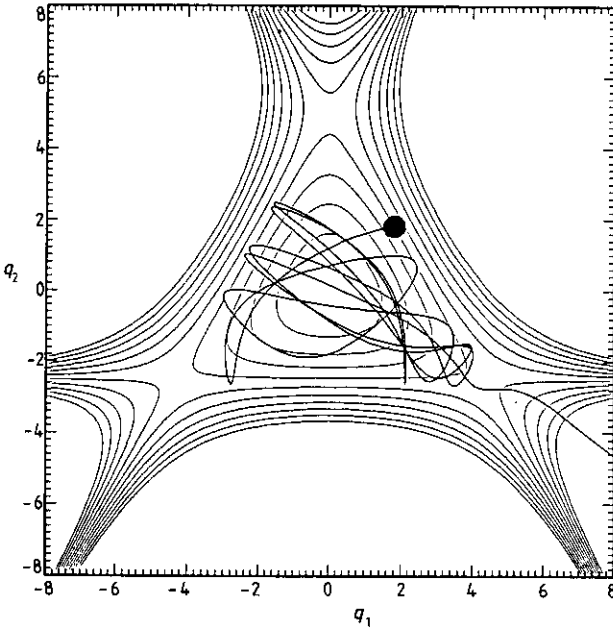


Figure 2. The same as figure 1 for the Hénon-Heiles system described by $\hat{H}_1(\hat{q}, \hat{p})$.

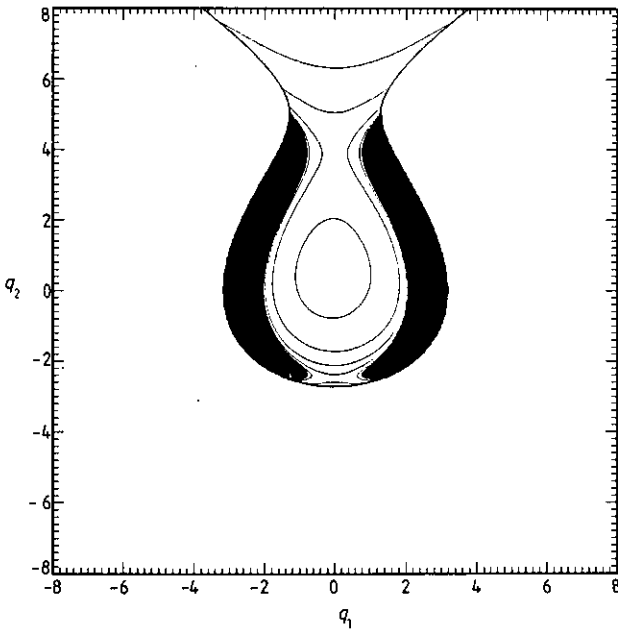


Figure 3. The distribution of the classical lifetimes for the separate system. The lifetime is defined as a time during which the particle is trapped in a circle with centre $(q_1, q_2) = (0, 0)$ and radius $r = 12$. Each initial condition has a common energy $E = 5.0$ and an incidence angle $\theta_{in} = \pi/6$ to q_1 -axis. The dark area represents the initial conditions with long lifetimes $t_s/2\pi (> 3.0)$.

in a scattering potential. As four independent parameters determining the initial condition, we choose q_1 , q_2 , the total energy, and the incidence angle to the potential. For the separable system, figure 3 shows the distribution of lifetimes of classical particles which start from the points distributed in the (q_1, q_2) plane. (The two remaining independent parameters are set to be constant.) In this figure, the time t_s during which the particle is trapped in the circle centred at $(q_1, q_2) = (0, 0)$ and with the radius $r = 12$ is represented by the contour lines from $t_s/2\pi = 0$ to $T_s (= 3.0)$ at an interval of 0.2. The dark area indicates the region satisfying the condition $t_s > T_s$.

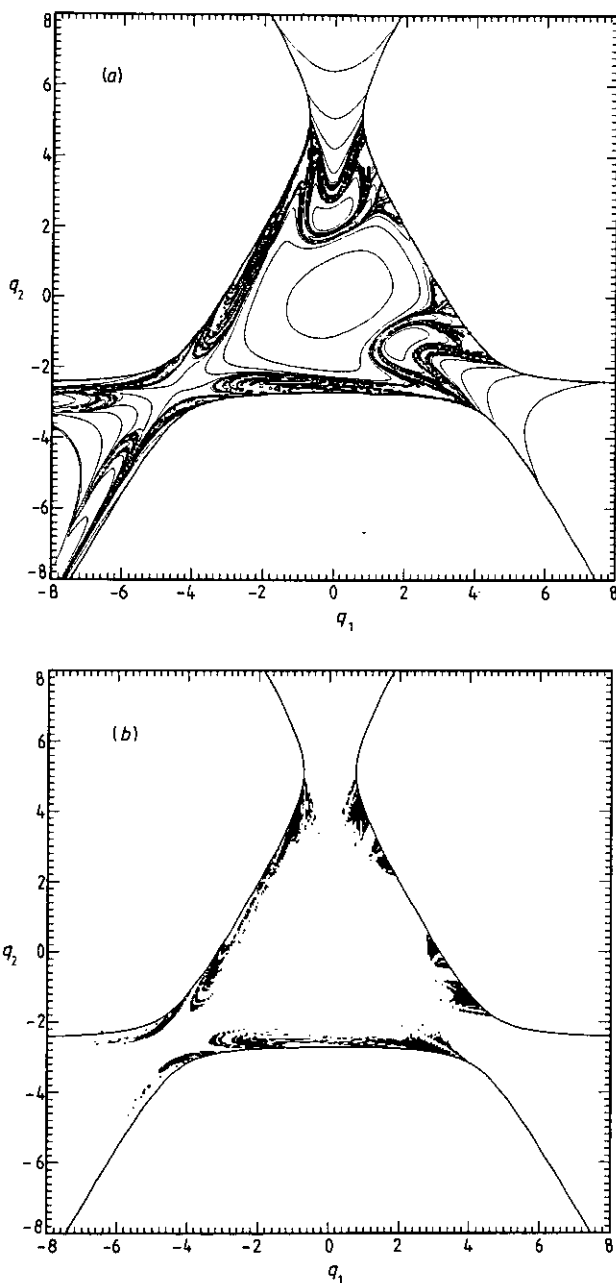


Figure 4. The same counter lines as figure 3 for the Hénon-Heiles system (a), and the regions of the initial conditions with long lifetimes (b).

This region occupies a large portion of the equi-energy space, and is hardly changed even if we increase T_s . These initial conditions provide periodic orbits, and have infinite lifetimes. (They construct an invariant set in the phase space.) In figure 4(a), the same contours as figure 3 are depicted for the coupled system. The regions satisfying the condition $t_s > T_s$ are depicted in figure 4(b). At this energy ($E = 5.0$),

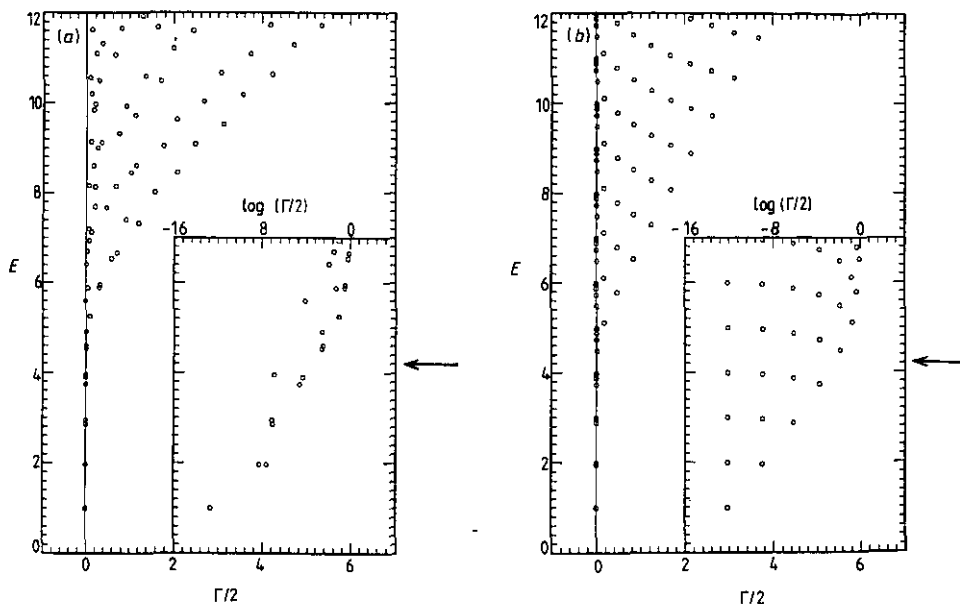


Figure 5. The distribution of the resonance eigenvalues of (a) $\hat{H}_1(\hat{q}, \hat{p})$ and (b) $\hat{H}_2(\hat{q}, \hat{p})$ on the complex energy plane. The arrow in each figure points to the height of the lowest barriers of the potential. For the energy region under the lowest barriers, the logarithmic plots are also shown in the inlets, since their imaginary parts are extremely small.

all the trajectories in the system would show the chaotic behaviour. Compared with the separable system, the pattern of the contour lines is highly complicated. The regions with long lifetimes are scattered and faint. (In contrast to the uncoupled system, this region becomes smaller if we increase T_s .) This means most of the particles rapidly escape from the circle. All of these characteristic features come from the chaotic behaviour of the classical particle in the non-separable system.

The global distribution of resonance eigenvalues for the two systems is given in figure 5. In the uncoupled system, the eigenvalues branch in a systematic manner. The lowest branch in figure 5(a) is the group of resonances derived from the scaling procedure for the q_2 -axis associated with the vibrational ground states of the q_1 -oscillator. Other higher branches may be associated with overtones. On the other hand, the coupling term of the Hénon–Heiles system destroys such regularity. Eigenvalues are rather randomly distributed for this case. This fact was also suggested by Seideman and Miller (1991) in conjunction with the transition state theory.

For the resonance eigenvalues under or near the potential barriers, Waite and Miller have already found a similar distribution due to the coupling between the two modes, which destroys the mode specificity of the unimolecular rate. They also investigated the one-barrier Hénon–Heiles system similar to our uncoupled system and showed the mode specificity. Then they concluded that the lack of the mode specificity is related to the three exit valleys. In such a low-energy region, although the rate constants due to tunnelling are extremely small (as is shown in the inlets in figure 5), they are meaningful in the transition state theory (Waite and Miller 1981, Seideman and Miller 1991). However, since under the barriers a quantal particle with a finite lifetime is different to a classical particle, the classical quasi-periodic/ergodic behaviour may not be related to the rate constants.

On the other hand, in the energy region over the barriers where the lifetimes of both particles are finite, we may compare the quantum dynamics with the classical one. When we investigate the distribution of resonance states with enough lifetimes to be observed, the total spectrum of the Hamiltonian plays an important role. The spectral density $\rho(E)$ is easily calculated from the distribution of resonance eigenvalues $\{\tilde{E}_n\}$:

$$\rho(E) = -\frac{1}{\pi} \text{Im} \lim_{\epsilon \rightarrow +0} \text{tr} \left[\frac{1}{E + i\epsilon - \hat{H}} \right] = -\frac{1}{\pi} \text{Im} \sum_n \frac{1}{E - \tilde{E}_n}. \quad (3.1)$$

Both of the two systems have many sharp resonances even in the high-energy region over the barriers (figure 6). This phenomenon occurs only in multi-dimensional potentials which have some infinite walls, and is not seen in one-dimensional systems. It should be noted that the sharp resonances are more prominent in the uncoupled system than in the Hénon–Heiles system. This fact shows that in quantum mechanics the particle in the non-integrable system is also less bounded than the particle in the integrable system.

The eigenstates of the uncoupled system (figure 5(b)) can be assigned as (n_1, n_2) using the vibrational quantum number n_1 and n_2 . The eigenstates with low values of n_1 near the real energy axis contain only the low-lying states of the q_2 mode, the coordinate of which extends toward the exit valley. Thus they have a very long lifetime and provide the spectrum with extremely sharp resonances, the width of which is attributed to a little tunnelling effect towards the q_2 -axis. It is natural to correlate them to classical quasi-periodic trajectories. They may be quantized by an appropriate quantization rule so as to give the corresponding eigenstates of the series. Other eigenstates with the higher values of n_2 have finite lifetimes proportional to $1/\Gamma$ and provide broader resonances. They can be correlated to the classical trajectories in

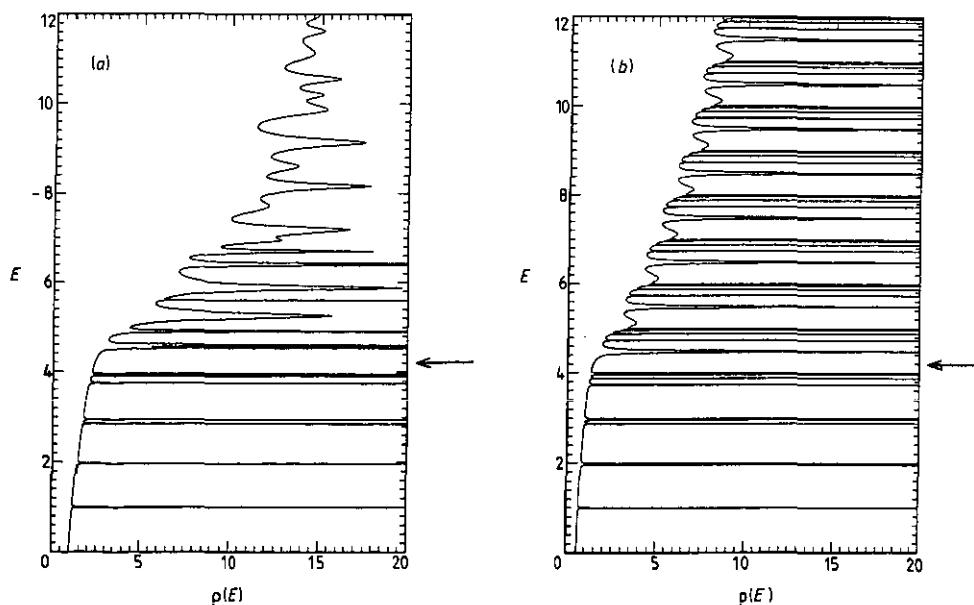


Figure 6. The continuous spectral density of (a) $\hat{H}_1(\hat{q}, \hat{p})$ and (b) $\hat{H}_2(\hat{q}, \hat{p})$ calculated from the resonance eigenvalues. The arrow in each figure points to the height of the lowest barriers of the potential.

which a classical particle quickly escapes from the central part of the system after few reflections on the wall.

In the spectrum of the Hénon–Heiles system, the extremely sharp resonances disappear in the energy region over the barriers (figure 6(a)). This corresponds to the fact that in classical mechanics no quasi-periodic motion (with infinite lifetime) is possible in this energy region. Some sharp (or little-broadened) states in this energy region may be related to the classical ergodic (chaotic or stochastic) motions which have comparatively long classical lifetimes because of repeated (irregular) reflections on the three walls. The number of these states are decreased with increasing energy in contrast to the case of the uncoupled system which has many extremely sharp resonances regardless of the energy. The more broad resonances, which may not be observed in this figure, correspond to the classical escape trajectories which immediately leave the central part of the potential. Of course, we cannot strictly distinguish these escape trajectories from the ergodic ones only by their lifetimes, because their lifetimes continuously join. To compare the classical and quantum systems in the more strict sense from the standpoint of the lifetime, we must consider the distribution of the classical and quantal life times at each energy.

In summary, we show the importance of getting the global distribution of resonances for the quantum dynamics of multi-dimensional systems. The global distribution including resonances far from the real axis clearly reflects the characteristics of a system. The spectral density also represents the quantum mechanical feature, associated with the classical behaviour.

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