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Classical Limit of Fredholm Theory for Elastic and Inelastic Scattering; Inability of Phase Space Integrals to Describe Inelastic Transitions

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Abstract

Using a classical phase space integral to approximate certain quantum mechanical operations, the classical limit of the Fredholm determinant for a general multichannel scattering system is derived. This statistical form of the classical limit of scattering is found to give exactly the same "classical S-matrix" for the case of potential scattering as does a dynamical treatment of the classical limit. For inelastic scattering, however, the classical S-matrix which results from this Fredholm determinant is only an approximation to that obtained by exact classical dynamics. Reasons for this failure of the statistical version of the classical limit for inelastic scattering are discussed.
I. INTRODUCTION

In two recent papers\textsuperscript{1,2} we have dealt with the classical-limit eigenvalue relation for general non-separable dynamical systems. A particularly interesting feature of this study of the bound-state problem is the correspondence between the dynamical form of the quantum condition and a statistical form. The dynamical version involves an action integral along a particular classical trajectory (the periodic trajectory of the system corresponding to a given energy $E$), whereas the statistical version is expressed in terms of a phase space integral (the volume of phase space with energy less than or equal to the given value $E$). The practical importance of this correspondence between statistical and dynamical approaches lies in the fact that statistical methods are generally much easier to apply than dynamics.

Analogous to these treatments of bound-state problems we explore in this present paper the relation between dynamical and statistical approaches to the classical limit of scattering for a general non-separable system. Thus it has previously been shown\textsuperscript{3} how one can use exact classical trajectories for a general collision system to construct the classical-limit of the quantum mechanical $S$-matrix (the "classical $S$-matrix") for the scattering processes. Corresponding to this dynamical prescription for obtaining the classical $S$-matrix, therefore, we wish to find the statistical procedure (i.e., one based on phase space integrals) which is related to it.
In order to use a formalism developed for bound-state problems, one can always convert a scattering problem into a bound-state one by some variation of "box normalization". The statistical version of the classical limit of scattering can then be obtained by invoking the statistical eigenvalue relation established previously. As a particularly simple example of how scattering is related to the box-normalized eigenvalue problem, consider s-wave potential scattering; the WKB (i.e., classical limit) phase shift can be written as

\[ \eta(E) = \int_{r_0}^{R} \, dr \, k(r) - \int_{0}^{r_0} \, dr \, k, \]

where \( k(r) = \left( \frac{2\mu(E-V(r))}{\hbar^2} \right)^{\frac{1}{2}}, \) \( k = \left( \frac{2\mu E}{\hbar^2} \right)^{\frac{1}{2}}, \) \( V(r) \) is the scattering potential, \( E \) the collision energy, \( \mu \) the reduced mass, \( r_0 \) the classical turning point (i.e., \( V(r_0) = E \)), and \( R \) is some indefinitely large value. Eq.(1) may be interpreted in terms of classical-limit eigenvalues: Thus \( n(E) \),

\[ \left[ n(E) + \frac{1}{2} \right] \pi = \int_{r_0}^{R} \, dr \, k(r), \]

is the classical-limit quantum number function [the inverse function of the eigenvalue function \( E(n) \)] for the "potential well" formed by the actual potential \( V(r) \) with an impenetrable barrier imposed at \( r=R \); Eq.(2) is the well-known Bohr-Sommerfeld quantum condition for this box-normalized potential. Similarly, \( n_o(E) \), the quantum number function for the potential well with \( V(r) \) replaced by \( 0 \), is defined by the relation in Eq.(2) with \( k(r) \) replaced by \( k \). Eq.(1) for the phase shift is thus written in terms of these quantum number functions as
\[ \eta(E) = \pi [u(E) - n_o(E)]. \] (3)

Scattering results are obtained, therefore, in terms of the eigenvalues of the box-normalized system and those of its unperturbed \((V=0)\) counterpart.

For more general collision systems (i.e., ones with internal degrees of freedom) it is also possible to extract the S-matrix from a consideration of the box-normalized eigenvalue problem; the theoretical machinery for doing this is Fredholm theory.\(^5\) [Although present day discussions of Fredholm theory do not resemble this box normalization approach, it originated from such considerations.\(^6\)] Rather than follow the box normalization procedure directly, therefore, we develop the classical phase space approximation for the Fredholm determinant and then appeal to the general results of Fredholm theory to construct the S-matrix. Section II discusses the classical limit of Fredholm theory as it applies to potential scattering, and Section III carries this out for the more general case of inelastic scattering.

This statistical version of the classical limit of Fredholm theory is seem to reproduce the usual classical S-matrix for the case of potential scattering, but it is unfortunately not able to provide the correct description of inelastic scattering. Reasons for this failure are discussed in Section IV.

**II. POTENTIAL SCATTERING**

For a general discussion of Fredholm theory (as it applies to scattering) the reader is referred to the text by Newton;\(^5\) a clear summary of the basic results is also contained in the
recent work by Reinhardt and co-workers who have shown that the Fredholm approach can also be a useful tool for quantum mechanical computations. In what follows we shall simply extract the results of the general theory which are required for our purposes.

The $S$-matrix for potential scattering (a one-dimensional matrix in this case) is given in terms of the Fredholm determinant $\Delta(k)$ by

$$ S = \frac{\Delta(-k)}{\Delta(k)}, $$

(4)

where $k = (2\mu E/h^2)^{1/2}$, $E$ being the collision energy; since

$$ \Delta(-k) = \Delta(k)^*, $$

(5)

one sees that $S$ is a complex number of unit modulus. The Fredholm determinant is in turn given by formal expression

$$ \Delta(k) = \det[(E-H)(E-H_0)^{-1}], $$

(6)

where $H = H_0 + V$ is the Hamiltonian operator for the system, and $V$ is the scattering interaction. By the determinant of an operator $A$ one means the determinant of its matrix representation in some complete set of states:

$$ \det[A] = \det|\langle i | A | j \rangle|. $$

(7)

To evaluate the determinant of an operator, it is convenient to employ the identity

$$ \det[A] = \exp[\text{tr}(\ln A)], $$

(8)

where $\text{tr}(\ln A)$ means the trace of the operator $\ln A$; i.e.,

$$ \text{tr}(\ln A) = \sum_i \langle i | \ln A | i \rangle, $$

(9)
where the sum is over some complete set of states. Combining
Eqs. (6) and (8), one has

\[ \ln \Lambda(k) = \text{tr}(\ln[(E-H)(E-H_0)^{-1}]). \quad (10) \]

All of the above equations of this Section are exact quantum
mechanical relations; the statistical version of the classical
limit approximation for the Fredholm determinant is obtained by
using a phase space integral to evaluate the trace in Eq. (10).
Thus if \( B_{op} \) is some operator which is expressed in terms of
the coordinate and momentum operators \( r_{op} \) and \( p_{op} \) as \( B(r_{op}, p_{op}) \),
then the phase space approximation for its trace is

\[ \text{tr}(B_{op}) = h^{-1} \int dr \int dp \ B(r,p), \quad (11) \]

where \( B(r,p) \) is the same function of the classical variables \( r \)
and \( p \) that \( B_{op} \) is of \( r_{op} \) and \( p_{op} \); \( h = 2\pi \hbar \) is Planck's constant.
Since all the operators of interest for our purposes are
Hamiltonians, there is no ambiguity about the appropriate
correspondence with the classical functions.

With the phase space approximation of Eq. (11), Eq. (10)
becomes

\[ \ln \Lambda(k) = h^{-1} \int_0^\infty dr \int_{-\infty}^{\infty} dp \ \{\ln[E-H(p,r)] - \ln[E-H_0(p,r)]\}, \quad (12) \]

and the classical Hamiltonian functions are

\[ H(p,r) = p^2/2\mu + V(r) \quad (13a) \]

\[ H_0(p,r) = p^2/2\mu ; \quad (13b) \]

the domain of integration is, as indicated, all of phase space.
The integral over momentum can be carried out by elementary
\[ \int_{-\infty}^{\infty} dp \ln(E-H) - \ln(E-H_0) = 2\pi i \hbar [k - k(r)], \]  

where \( k = (2\mu E/\hbar^2)^{1/2} \) and \( k(r) = [2\mu (E-V(r))/\hbar^2]^{1/2} \); if \( E - V(r) < 0 \), then \( k(r) = i|k(r)| \). Eq. (12) thus becomes

\[ \ln \Delta(k) = -i \int_0^\infty dr k(r) - k, \]  

or

\[ \Delta(k) = \exp(\theta - i\eta) \]  

where

\[ \theta = \int_{r_0}^R dr |k(r)| \]

\[ \eta = \lim_{R \to \infty} \int_{r_0}^R dr k(r) - \int_{r_0}^0 dr k, \]

\( r_0 \) being the classical turning point; i.e., \( \theta \) is real, and \( \eta \) is the WKB phase shift.

With the Fredholm determinant given by Eq. (16), Eq. (14) then gives the S-matrix as

\[ S = \exp(2i\eta^{\text{WKB}}). \]  

This is the usual result which is also obtained by a strictly dynamical approach.

Before concluding this discussion of potential scattering, it is useful to note a few details of the replacement of \( k \) by \( -k \) in applying Eq. (4) to the phase integral in Eqs. (15) and (16). In classically allowed regions \((r > r_0)\) one has
\[ k(r) \rightarrow -k(r) \]

when \( k \) is replaced by \(-k\); in classically forbidden regions \((r<r_0)\), however, one has

\[
-ik(r) \equiv |k(r)| \rightarrow |k(r)| \equiv -ik(r).
\]

With regard to \( k + -k \), therefore, the real (imaginary) part of \( k(r) \) is considered to be an odd (even) function of \( k \). For this reason the contribution to the coordinate integral in Eq.(15) from classically forbidden regions cancels out in the ratio in Eq.(4).

**III. INELASTIC SCATTERING**

Now suppose there are internal degrees of freedom in addition to the scattering (translational) degree of freedom. The internal degrees of freedom are quantized in the initial and final asymptotic regions, and the S-matrix is the matrix of transition probability amplitudes from initial internal states (or channels) to final internal states (or channels); the dimension of the S-matrix is the number of energetically accessible internal states (the number of open channels).

The Fredholm determinant is still given formally by Eq.(6), but one must now consider it to be an independent function (sign-wise at least) of all the channel momenta; i.e.,

\[
\Delta(k) \equiv \Delta(k_1,k_2,\ldots) = \exp \left\{ \text{tr}\left[ \ln(E-H)(E-H_0)^{-1} \right] \right\}, \quad (18)
\]

where the asymptotic momentum (in units of \( \hbar \)) for channel \( i \) is

\[
k_i = \left[ 2\mu(E-E_i)/\hbar^2 \right]^\frac{1}{2},
\]
E being the (fixed) total energy and $c_i$ the internal energy of internal state $i$. The channel momenta $\{k_i\}$ are all related to one another through their definition in Eq. (19), and the only sense in which they are considered independent in Eq. (18) is that one needs to change the sign of some of them and not to change the sign of others.

The $S$-matrix is given in terms of the Fredholm determinant of Eq. (18) by

$$S_{i,i} = \frac{\Delta_i(k)}{\Delta(k)}$$

$$S_{i,j} = [S_{i,i} S_{j,j} - \Delta_{i,j}(k)/\Delta(k)]^{\frac{1}{2}},$$

where $i \neq j$, and

$$\Delta_i(k) = \Delta(k_1, k_2, \ldots, -k_i, \ldots)$$

$$\Delta_{i,j}(k) = \Delta(k_1, k_2, \ldots, -k_i, \ldots, -k_j, \ldots).$$

As in the preceding Section we use a classical phase space integral to calculate $\Delta(k)$ and then construct the $S$-matrix by using Eqs. (20) and (21).

For notational convenience we assume there to be just one internal degree of freedom; the treatment is identical for any number of internal degrees of freedom. With the phase space integral approximation to the trace in Eq. (18), one has

$$\ln \Delta(k) = h^{-2} \int_0^{2\pi} dq \int_0^\infty dn \int_0^\infty dr \int_{-\infty}^\infty dp [\ln(E-H) - \ln(E-H_0)],$$

where the normalization factor is $h^{-2}$ since there are two degrees of freedom. Since an integral over all phase space is
CONTINUE

312 FORMAT (21X, *INTENSITY AS A FUNCTION OF WAVELENGTH WITH FILM THICKNESS AS A PARAMETER ON AN ELECTRODE, 

5 FORMAT (10X, *WAVELENGTH* 4X, 9F11.4)
6 FORMAT (10X, *PHASE CHANGE* 10X, *METAL* 9X, 9F11.4)
7 FORMAT (10X, *FILM THICKNESS* 45X, *INTENSITY*/

11 FORMAT (13X, 14, 7X, 9F11.4)
13 FORMAT (10X, *TOTAL* 9X, 9F11.2, 

STOP
END

FUNCTION RIC(WAVLTH)

THIS FUNCTION GIVES REFRACTIVE INDEX OF THE SOLUTION AT A SPECIFIED WAVELENGTH USING A POLYNOMIAL OF 6 TERMS.

COMMON /RICOM/ B,NO
DIMENSION B(20,8)
REAL A
A=WAVLTH
C=0.0
A=A/4000
DO 601 K=1,NO
C=C+B(K)*A**(K-1)
601 CONTINUE
RIC=C
RETURN
END

FUNCTION REFRXC (SCALE, LAMBDA)

THIS FUNCTION GIVES REFRACTIVE INDEX OF THE SOLUTION FROM THE SCALE READINGS OF THE REFRACTOMETER.

FOR THE NEW PRISM (749-1)

REAL NPRIS, PC(8), LAMB
LAMB=LAMBDA/4000
APRIS=0.0174533*68,0
PC(1)=2.109825761
PC(2)=1.8465729697
PC(3)=2.4203415165
PC(4)=-1.6347815115
PC(5)=0.5602598970
PC(6)=-0.0773841103
NPRIS=0.
DO 701 J=1,6
NPRIS=NPRIS+PC(J)*(LAMB**((J-1))
701 CONTINUE
AMEAS=24.0-(2.0*SCALE)/3.0
AMEAS=AMEAS*0.0174533
ACALC=ASIN(SIN(AMEAS)/NPRIS)
APRIM=APRIS-ACALC
REFRXC=SiN(APRIM)*NPRIS
RETURN
END
MAXIMA AND MINIMA OF INTENSITY AS A FUNCTION OF WAVELENGTH ON A PLATINUM ELECTRODE

S POLARIZATION

WAVELENGTH  (NM)  FILM THICKNESS  (NM)  INTENSITY

390  NM  MAXIMA AT  30  NM  217  8563
405  492  779  964  1154  1341
MINIMA AT  120  NM  1699
327  495  642  869  1056  1244  1431

395  NM  MAXIMA AT  30  NM  8567
220  409  599  769  979  1168  1358
MINIMA AT  120  NM  1720
310  499  693  879  1069  1259  1449

390  NM  MAXIMA AT  30  NM  8571
222  414  607  799  991  1183  1375
MINIMA AT  120  NM  1774
312  504  697  889  1081  1273  1465

395  NM  MAXIMA AT  30  NM  8574
225
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independent of the particular canonical variables one uses to carry out the integral, in Eq. (22) we have chosen the usual translational coordinate and momentum \((r,p)\) for the translational degree of freedom, but have used the action-angle variables \(\{(\hbar n,\mathbf{q})\}\) for the internal degree of freedom. The factor \(\hbar\) has been included explicitly in the definition of the action variable, so that the quantity \(n\) is dimensionless and is the classical equivalent of the quantum number of the internal degree of freedom. In terms of these canonical variables the Hamiltonian functions are

\[
H = \frac{p^2}{2\mu} + \epsilon(n) + V(r,q,n),
\]

\[
H_0 = \frac{p^2}{2\mu} + \epsilon(n),
\]

where \(\epsilon(n)\) is the eigenvalue function for the isolated internal degree of freedom, and \(V\) is the scattering interaction.

Just as in the case of potential scattering, the integral over the translational momentum can be carried out by elementary means, and upon doing this Eq. (22) becomes

\[
\ln \Delta(\mathbf{k}) = -i \int_0^\infty dn (2\pi)^{-1} \int_0^{2\pi} dq \int_0^\infty dr \ k(r,q,n) - k(n), \quad (23)
\]

where the momenta are defined as

\[
k(n) = \left\{2\mu[E - \epsilon(n)]/\hbar^2\right\}^{\frac{1}{2}}
\]

\[
k(r,q,n) = \left\{2\mu[E - \epsilon(n) - V(r,q,n)]/\hbar^2\right\}^{\frac{1}{2}}.
\]

Classically, of course, \(n\) is a continuous variable, whereas quantum mechanically it is quantized. To make the appropriate identification with the discrete internal states, therefore, we...
make the replacement

$$\int_{0}^{\infty} dn = \sum_{n}$$

(24)

where the sum is over all integer values of \(n\). Eq.(23) thus becomes

$$\ln \Delta(k) = -i \sum n (2\pi)^{-1} \int_{0}^{2\pi} dq \int_{0}^{\infty} dr \, k(r, q, n) - k(n).$$

(25)

One may further simplify matters by noting that, just as in the previous section, the energetically forbidden regions of phase space in the integrals in Eq.(25) will cancel when one constructs the ratios of Fredholm determinants in Eq.(20). This follows from the discussion at the end of Section II. This means that Eq.(25) is of the form

$$\ln \Delta(k) = E(k) - i \sum n \eta(k_n),$$

(26)

where \(E(k)\) is real and an even function of all the channel momenta, and

$$\eta(k_n) = (2\pi)^{-1} \int_{0}^{2\pi} dq \lim_{R \to \infty} \left[ \int_{R}^{R} dr \, k(r, q, n) - \int_{0}^{R} dr \, k(n) \right];$$

(27)

\(\eta(k_n)\) is an odd function of \(k_n\) and is seen to be the WKB phase shift for the frozen internal degree of freedom (i.e., fixed \(n\) and \(q\)), which is then averaged over the angle variable \(q\).

From Eq.(26) for the Fredholm determinant, it is easy to see that the determinants in Eq.(21) are given by

$$\Delta_i(k) = \Delta(k) \exp[2i\eta(k_i)]$$

(28a)

$$\Delta_{i,j}(k) = \Delta(k) \exp[2i\eta(k_i) + 2i\eta(k_j)],$$

(28b)
so that the S-matrix elements of Eq. (20) are

\[ S_{i,i} = \exp[2i\pi(k_i')] \]  

(29a)

\[ S_{i,j} = 0, \ i \neq j. \]  

(29b)

There are no inelastic transitions in this approach, therefore, and the elastic scattering is only an approximation to the exact classical dynamics. Eq. (29a) is, in fact, a classical-limit version of the sudden approximation for the elastic scattering in channel i.

IV. DISCUSSION

The shortcomings of this statistical version of the classical-limit of inelastic scattering are probably most directly related to the asymptotic degeneracy that is inherent in a multi-channel scattering system. Thus in establishing the correspondence between statistical and dynamical quantum conditions it was essential that there was only one periodic trajectory of the system for a given energy. If other periodic trajectories existed, they had to be related to some constant of the motion (such as total angular momentum) or a discrete symmetry of the system and explicitly removed; the eigenvalue problem could then be considered separately for each value of the conserved quantity or discrete symmetry, there then being only one periodic trajectory of the system at the given energy for that particular subspace.

For a multi-channel scattering system, however, there are \( N (N \equiv \text{number of open channels}) \) degenerate quantum states of the system, corresponding to the \( N \) different internal states
which can be the initial state. When the system is enclosed in a finite box, these \( N \) degenerate states are split—only in the limit of an infinite box does the degeneracy appear. Furthermore, this degeneracy is not related to any constant of motion or discrete symmetry—rather it is associated with constants of the motion of the unperturbed Hamiltonian to and from which the system evolves asymptotically.

Corresponding to this quantum degeneracy, there is thus more than one "periodic" classical trajectory for the system at a given energy. [Periodicity comes about here only artificially by reflection from the walls of the large "box"; the appropriate trajectories are actually the aperiodic scattering trajectories.] Similarly, these several "periodic" trajectories cannot be classified according to any discrete or continuous symmetry of the system; i.e., there is no way to decompose (or factor) the dynamical problem so that there is only one "periodic" trajectory per energy per subspace. As noted above, this difficulty is directly related to the asymptotic degeneracy that is an intrinsic feature of scattering systems with internal degrees of freedom; for in a bound state situation degeneracies can always be related to some symmetry of the system and thus explicitly removed (i.e., there are no accidental degeneracies).

One might imagine classifying the various "periodic" classical trajectories of the scattering system by the "constants of the motion" which are simply the initial conditions of the individual trajectories. Initial conditions, however, although they are constants of integration of the equations of motion, are not integral constants of the motion in the usual sense
functions of the coordinates and momenta— but not involving the time explicitly—which remain constant in time). Such an approach, too, introduces dynamics per se and clearly foils the attempt to develop a theory based solely on phase space integrals.

Acknowledgement

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*Alfred P. Sloan Fellow.


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