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Transition state theory for ballistic electrons

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Abstract. Transition state theory as developed for chemical reactions is transferred to ballistic electrons. The phase-space flux through the transition state is related to the conductance. Steps in the conductance follow from a semiclassical extension of this formula. Tunnelling corrections have to be included to yield the rounding of the steps. A magnetic field causes a sharpening of the steps and finite temperature an additional broadening. The calculation also shed some light on the abilities and limitations of the semiclassical approach.

1. Introduction

Investigations of two-dimensional ballistic electron gases have revealed a number of phenomena on the borderline between classical and quantum mechanics. Among them are the focusing of electrons in magnetic fields [1], conductance fluctuations [2], quantization of conductance [3, 4], non-additivity of resistance [5, 6] and periodic orbit modulations in magnetoresistance [7]. Since the dimensions of the systems are often just a few de Broglie wavelengths they are natural testing grounds for concepts in semiclassical physics. Because of the relationship between the scattering matrix and the Landauer–Büttiker formula for transmission [8, 9] considerable attention has been paid to the statistical properties of the S-matrix [2, 10], especially in the case of irregular scattering [11].

The models used for transmission of electrons through a channel have a close resemblance to models of reactive scattering in chemistry [12, 13]. The equivalent of a reaction is an electron passing through the channel. Most studies in the chemical physics literature have focused on one-dimensional channels in two-degree-of-freedom systems. In the case of chemical reaction models this restriction applies to collinear three-particle geometries only, but for ballistic electrons it is correct within the independent electron picture due to the lateral confinement. The ballistic electron models thus provide a rather direct illustration of ideas developed earlier [12, 13].

The aim of this paper is to translate the formalism of transition state theory to electron conductance and to derive a formula for the transmission through a channel. In the case of a channel with a single constriction this formula can be evaluated to three levels of approximation: classically, to leading-order in a stationary phase approximation and to higher-order including tunnelling corrections. In the latter case we can improve a formula first obtained by Miller [14] for the positions and the widths of the resonances in scattering. The present investigation will also shed some light on the accuracy with which semiclassical methods can be expected to work in the case of ballistic conductance.

The outline of the paper is as follows. In section 2 conductance will be related to classical fluxes and phase-space densities of states, thus preparing for the direct semiclassical

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analysis of section 3. The extension to a channel in a magnetic field is discussed in section 4. Some final comments, in particular on cases with irregular scattering, are given in section 5.

2. Classical transmission

Consider the idealized geometry for a transmission measurement shown in figure 1. Two electron reservoirs at chemical potential μ_L and μ_R are connected by a two-dimensional narrow wire with a constriction. It is assumed that the mean free path of the electron is larger than the size of the wire, and reservoirs do not reflect electrons before they are thermalized. Because of the difference in chemical potential there is a flux of electrons across the wire, inhibited only by the current-carrying capacity of the constriction. The ratio of the current, given by the flux multiplied by the charge, to the difference in potential is the conductance, or, more precisely since no dissipation is involved, the transmission.

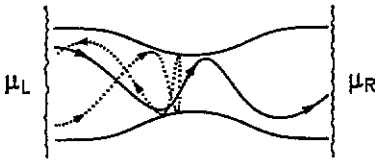

Figure 1. Transmission through a channel. Electrons move through the channels as idealized mass points, free of inelastic scattering. A constriction is modelled by a narrowing of the equi-energy contours. Some electrons pass through the constriction (full curve) and some are reflected back (broken curve). Far from the constriction, inelastic scattering equilibrates the electrons to the chemical potentials μ_R and μ_L of the reservoirs.

To determine the electrical current we need the number of electrons crossing from one side of the constriction to the other. To achieve this we count the electrons crossing a line $x = x_0$. Allowing for different momenta perpendicular to this line, the number of electrons at a fixed energy is obtained by integrating the phase-space flux density $\delta(x - x_0)v_x = \delta(x - x_0)p_x/m$ over all positions and momenta:

$$F(E) = \int \mathrm{d}x\,\mathrm{d}y\,\mathrm{d}p_x\,\mathrm{d}p_y\,\delta(E - H)\delta(x - x_0)\frac{p_x}{m}\Theta(\pm p_x)\chi(x, y, p_x, p_y). \tag{1}$$

The step function Θ selects the direction of the current; both signs yield the same value. The characteristic function χ selects the trajectories contributing to the current: it equals one on all trajectories which live on different sides of the channel for $t \to \pm\infty$, and zero for the others.

The rate of transition from one side of the channel to the other is obtained by integrating the product of $F(E)$ with the density of states and the occupation probability over all energies. Here the density of states is $2/h^2$, because there are two spin directions per quantum cell of size h^2. The occupation probability is given by the Fermi–Dirac statistics at temperature T and chemical potential μ:

$$f(E, \mu) = \frac{1}{\mathrm{e}^{(E-\mu)/kT} + 1}. \tag{2}$$

Thus, the transition rates become

$$w(\mu) = \int \mathrm{d}E\,\frac{2}{h^2} f(E, \mu)F(E). \tag{3}$$

Electrons can cross the channel in both directions. However, since there are different chemical potentials on the sides μ_L and μ_R respectively, these fluxes do not cancel. The

observed current is the difference in the rates of electrons going from left to right and from right to left, multiplied by the charge e:

$$I = e(w(\mu_L) - w(\mu_R)). \tag{4}$$

Expanding around the Fermi energy $E_F = (\mu_L + \mu_R)/2$ to first order in the differences of the chemical potentials and taking into account that this difference is charge multiplied by electrical potential drop, $\mu_L - \mu_R = eU$, one finds for the conductance that

$$g = \frac{I}{U} = \frac{2e^2}{h} \int dE \left(-\frac{\partial f}{\partial E}\right)_{\mu=E_F} \frac{F(E)}{h}. \tag{5}$$

For very low temperature the derivative of the Fermi–Dirac distribution becomes a δ-function so that the conductance is related to the microcanonical flux at the Fermi energy:

$$g|_{T=0} = \frac{2e^2}{h} \frac{F(E_F)}{h}. \tag{6}$$

The quotient $F(E)/h$ is dimensionless and counts the number of open channels by an adiabatic approximation.

3. Transition states

In the above formula we determine the current through the plane $x = 0$ in phase space. Actually, the formula holds true for any surface which divides phase space, and the step function selects the required electron trajectories, asymptotically crossing the barrier. If we wish to approximate the integral by eliminating the step function then, of course, a good choice of the surface becomes mandatory [15]. Evidently, one would like to minimize the number of backscattered trajectories (see figure 1). Surfaces which minimize the transmission are called transition states.

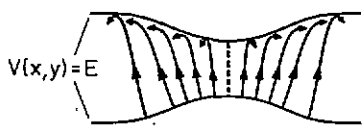

Figure 2. Periodic orbit dividing surface. As explained in the text every dividing surface across the channel has a point on the boundary $V(x) = E$ of the classically allowed region. Moving along the boundary, there is (in this geometry) exactly one orbit that connects the two boundaries (broken curve).

As the many detailed studies of Pollak, Pechukas and Child [16, 17] have shown, a good choice of surface of section is a periodic orbit dividing surface (PODS), i.e. a surface spanned by a trajectory connecting the equipotential boundaries. In brief, the idea is as follows. At the equi-energy contours, phase space becomes one-dimensional since all energy is absorbed by the potential energy and all momenta vanish. Thus, every dividing surface must have points on the equi-energy contours. Furthermore, the surface should be spanned by an orbit since this eliminates the possibility of trajectories being tangential to it. As shown in figure 2 for the geometry of figure 1, all trajectories started at points along the boundary will leave through a point on the other side, except for the one in the centre; this is the only orbit connecting both equipotential contours and the motion along it is periodic. Since motion along all classical orbits connecting two equi-energy contours is periodic, these transition

states are also called PODS. In the particular case of figure 2, the PODS lives along one of the coordinate axes and the integral for $F(E)$ can be performed easily:

$$F(E) = \int \mathrm{d}y\,\mathrm{d}p_y\,\Theta(E - H(x = x_0, p_x = 0, y, p_y)) = \oint p\,\mathrm{d}q. \tag{7}$$

The second form of the integral is just the action of the periodic orbit; this clearly gives a flux smoothly increasing with energy.

If there are several transition states, then the one with minimal action is a minimal upper bound on the flux crossing the channel. If there is just one surface, then it is not just an upper bound but is exact [15, 16].

4. Quantization of the conductance

Since the action of a periodic orbit in a smooth potential changes continuously with energy, the preceding calculations do not give steps in the conductance. To find these, one has to improve the density of states and take into account modulations due to classical periodic orbits [18]. Therefore, the local phase-space density $\rho_{kl}(p, x) = \delta(E - H(p, x))$ of equation (1) has to be replaced by the corresponding quantum mechanical expression which may be obtained from a Wigner transform of Green's function:

$$\rho_{qm}(p, x) = \int \mathrm{d}x'\,\mathrm{e}^{-\mathrm{i}px'/\hbar}\left\{-\frac{1}{\pi}\operatorname{Im} G(x + x'/2, x - x'/2; E)\right\}. \tag{8}$$

In this formula and in many of the others below, taking the imaginary part is a reminder to include a small imaginary part to the energy $E = E' + \mathrm{i}\epsilon$ and to take the limit of vanishing ϵ. The usual representation of the density of states in energy $\delta(E - H) = -\operatorname{Im}\operatorname{Tr} G/\pi$ can be obtained by integrating over all positions and momenta and dividing by h^2 (the phase-space volume of one state).

When substituted in the phase-space flux (1), one finds that for a transition state at $x_0 = 0$

$$\frac{F(E)}{h} = -\frac{1}{\pi}\operatorname{Im}\int \mathrm{d}p\,\mathrm{d}x\,\mathrm{d}x'\,\frac{1}{h}\frac{p_x}{m}\delta(x)\mathrm{e}^{-\mathrm{i}px'/\hbar}G(x + x'/2, x - x'/2; E). \tag{9}$$

Performing the integrations on x, p_y and y, one finds

$$\frac{F(E)}{h} = -\frac{1}{\pi}\operatorname{Im}\int \mathrm{d}p_x\,\mathrm{d}y\,\mathrm{d}x'\,\frac{p_x}{m}\mathrm{e}^{-\mathrm{i}px'/\hbar}G\left((x'/2, y), (-x'/2, y); E\right). \tag{10}$$

A semiclassical expression for Green's function $G(x_2, x_1; E)$ can be obtained from

$$G(x_2, x_1; E) = \frac{1}{\mathrm{i}\hbar}\int_0^\infty \mathrm{d}t\,\langle x_2|\mathrm{e}^{-\mathrm{i}Ht/\hbar}|x_1\rangle\mathrm{e}^{\mathrm{i}Et/\hbar} \tag{11}$$

by approximating the propagator by classical trajectories travelling from x_1 to x_2 in time t:

$$\langle x_2|\mathrm{e}^{-\mathrm{i}Ht/\hbar}|x_1\rangle = \sum_{\text{orbits}}\frac{1}{2\pi\mathrm{i}\hbar}|D_W|^{1/2}\mathrm{e}^{\mathrm{i}W/\hbar - \mathrm{i}\mu\pi/2}. \tag{12}$$

Here, W is the Lagrangian action

$$W = \int_0^t L(x, \dot{x})\,\mathrm{d}t = \int_{x_1}^{x_2} p\,\mathrm{d}x - Et \tag{13}$$

(with L Lagrange's function) which may also be expressed in terms of Hamilton's action $S = \int p\mathrm{d}q$ and the energy E at which the trajectory runs. D_W is the matrix of second derivatives

$$D_W = \det\left|\frac{\partial^2 W}{\partial x_2 \partial x_1}\right| \tag{14}$$

and μ counts the number of caustics encountered along the path.

The integrals that appear when these expressions are substituted in equation (10) are evaluated in stationary phase. As a function of x' the phase is stationary for $x' = 0$, and as a function of y one finds the condition that the initial and final y-components of the momentum must coincide:

$$\frac{\partial}{\partial y} S((0, y), (0, y)) = p_y|_{\text{initial}} - p_y|_{\text{final}} = 0. \tag{15}$$

This is just the condition for periodic motion in the y-direction at $x = 0$, i.e. at the transition state.

In the neighbourhood of the trajectory for small x', one can connect the action $S((-x'/2, y), (x'/2, y))$ with the linearization of motion perpendicular to the trajectory [18]. A small deviation $\delta z = (\delta x, \delta p_x)$ perpendicular to the trajectory will be mapped after one period T_y into $\delta z' = M\delta z$ with a symplectic 2×2 matrix M. The eigenvalues $\mathrm{e}^{\pm u}$ of this matrix are independent of the position along the orbit and are real for unstable orbits. If one factorizes the stability exponent into $u = \Lambda T_y$ with T_y the period and Λ the Lyapunov exponent for motion in the y-direction, then one can approximate the motion in x by that over an inverted parabola with effective Hamiltonian $\tilde{H} = (\tilde{p}_x^2 - \Lambda^2 \tilde{x}^2)/2$. This correctly describes the action perpendicular to the trajectory up to second order in the deviations. Since the propagator only contains the actions, the precise form of $(\tilde{p}, \tilde{x})$ and of the Hamiltonian $\tilde{H}$ are irrelevant. We thus have the important result that in the neighbourhood of the trajectory, motion is separable into a periodic motion and an unstable motion perpendicular to it. It is this separability that allows one to proceed further along the lines of Gutzwiller [18].

For separable motions the propagator factorizes and Green's function becomes a convolution of the Green functions G_x and G_y for motion in the x and y directions:

$$\begin{aligned} G(x_2, x_1; E) &= \frac{1}{\mathrm{i}\hbar}\int_0^\infty \mathrm{d}t\, \langle x_2|\mathrm{e}^{-\mathrm{i}Ht/\hbar}|x_1\rangle \mathrm{e}^{\mathrm{i}Et/\hbar} \\ &= \int \mathrm{d}E_x\, G_x(x_2, x_1; E_x) G_y(y_2, y_1; E - E_x). \end{aligned} \tag{16}$$

Upon substitution into (10) one obtains a convolution of the density of states at the transition state and a flux D perpendicular to it:

$$\frac{F(E)}{h} = \int \mathrm{d}E_x\, \rho_y(E - E_x) D(E_x) \tag{17}$$

with

$$\rho_y(E) = -\frac{1}{\pi}\,\mathrm{Im}\,\mathrm{Tr}\, G_y \tag{18}$$

and

$$D(E) = \int \mathrm{d}p_x\, \mathrm{d}x'\, \frac{p_x}{m} G_x(-x'/2, x'/2; E)\mathrm{e}^{-\mathrm{i}p_x x'/\hbar}. \tag{19}$$

As long as only real trajectories are admitted, the flux across a 1D barrier, as represented by the inverted barrier, equals one if the energy is above the barrier and zero otherwise. One is thus left with an integral over the density of states in the transition state:

$$\rho_y(E) = -\frac{1}{\pi}\,\mathrm{Im}\,\frac{1}{\mathrm{i}\hbar}\int \mathrm{d}y \int_0^\infty \mathrm{d}t\,\langle y|\mathrm{e}^{-\mathrm{i}H_y t/\hbar}|y\rangle \mathrm{e}^{\mathrm{i}Et/\hbar}. \tag{20}$$

For short times one has to substitute the exact propagator which yields the mean density of states (also known as the Thomas–Fermi term):

$$\rho_0(E) = \frac{T_y(E)}{h} \tag{21}$$

where T_y is the period of the y-motion. The periodic motion contributes at every return, whence

$$\begin{aligned}\rho_y(E) - \rho_0(E) &= +\frac{1}{\pi}\,\mathrm{Im}\sum_{r=1}^{\infty}\frac{\mathrm{i}T_y}{\hbar}\mathrm{e}^{(\mathrm{i}S_y/\hbar - \mathrm{i}\nu_y\pi/2)r}\\ &= +\frac{1}{\pi}\,\mathrm{Im}\,\frac{\mathrm{i}T_y}{\hbar}\frac{\mathrm{e}^{\mathrm{i}S_y/\hbar - \mathrm{i}\nu_y\pi/2}}{1-\mathrm{e}^{\mathrm{i}S_y\hbar - \mathrm{i}\nu_y\pi/2}}\\ &= -\frac{1}{\pi}\,\mathrm{Im}\,\frac{\partial}{\partial E}\log\left(1-\mathrm{e}^{\mathrm{i}S_y/\hbar - \mathrm{i}\nu_y\pi/2}\right).\end{aligned} \tag{22}$$

Expressing ρ_0 in the form

$$\rho_0 = -\frac{1}{\pi}\,\mathrm{Im}\,\frac{\partial}{\partial E}\log \mathrm{e}^{-\mathrm{i}\pi S_y(E)/h} \tag{23}$$

one can combine both expressions to find

$$\rho_y(E) = -\frac{1}{\pi}\,\mathrm{Im}\,\frac{\partial}{\partial E}\log\left(\mathrm{e}^{-\mathrm{i}\pi S_y(E)/h} - \mathrm{e}^{\mathrm{i}\pi S_y(E)/h - \mathrm{i}\nu\pi/2}\right). \tag{24}$$

In evaluating this formula one again has to keep in mind that a small positive imaginary part ϵ has to be added to the energy and that the limit of vanishing ϵ has to be taken. The flux $F(E)/h$ thus equals the total number of states above the barrier [19, 20]:

$$\frac{F(E)}{h} = -\frac{1}{\pi}\,\mathrm{Im}\log\left(\mathrm{e}^{-\mathrm{i}\pi S_y(E)/h} - \mathrm{e}^{\mathrm{i}\pi S_y(E)/h - \mathrm{i}\nu\pi/2}\right). \tag{25}$$

In smooth potentials with two turning points the Maslov index is $\nu = 2$ so that the argument of the logarithm becomes $2\cos(\pi S_y(E)/h)$ and real, and so $F(E) =$ constant. However, at the zeros of the cosine the phase jumps by π, resulting in jumps of $F(E)/h$ by one. The zeros of the cosine are given by the semiclassical quantization rules for periodic motion on the transition state, $S_y(E_n) = (n+1/2)h$. If $\Theta(E)$ denotes the step function one can finally write

$$g = \frac{2e^2}{h}\sum_n \Theta(E - E_n). \tag{26}$$

Thus, the conductance increases in steps whenever a new state fits into the transition state. According to this theory the steps are sharp in the microcanonical representation, the only possible source of smoothing being temperature effects in the average (5).

A quantum mechanical rounding of the steps at fixed energy arises from tunnelling corrections. Semiclassically, this requires inclusion of complex trajectories. For an inverted parabola $-\Lambda^2 x^2/2$ with maximum at $E = 0$, the flux then becomes

$$D(E) = \frac{1}{1+\mathrm{e}^{-\sigma(E)}} \tag{27}$$

with

$$\sigma(E) = \frac{2\pi E}{\hbar\Lambda}. \tag{28}$$

This is very small for $2\pi E < \hbar\Lambda$ and approaches one for $2\pi E > \hbar\Lambda$. Convoluted with the density of states in y this means that every step $\Theta(E - E_n)$ has to be replaced by $D(E - E_n)$.

When separating the propagator for the x- and y-motion it was tacitly assumed that the instability of the trajectory did not change with energy, i.e. the x-motion was independent of the y-motion; however, as the preceding discussion shows, only a small interval of the energy around the threshold E_n is important. It is, therefore, reasonable to substitute the Lyapunov exponent at that particular energy. One then arrives at the formula for the conductance in a smooth potential with two turning points:

$$g = \frac{2e^2}{h} \sum_n \frac{1}{1 + \exp(-2\pi(E - E_n)/\hbar\Lambda(E_n))} \tag{29}$$

together with the semiclassical quantization condition for the transition state:

$$S_y(E_n) = (n + 1/2)h. \tag{30}$$

This kind of problem was previously considered by Miller [14]. However, he does not include the mean density of states in his discussion and, furthermore, arrives at a result with approximations interchanged: the harmonic approximation is applied to the motion along the transition state, whereas the full tunnelling integral is taken. Both forms are useful depending on the rate with which the tunnelling integral and the action of the transition state are changing; the one deviating most from a harmonic approximation should be kept. For instance, in the case of the ballistic channel the tunnelling through the barrier may be approximated harmonically if the constriction is sufficiently smooth. The steps are then sufficiently sharp to be isolated and the non-harmonicity of the transition state may be detected.

The typical qualitative behaviour of the conductance is illustrated by a harmonic barrier with Hamilton function

$$H(p, x) = \tfrac{1}{2}(p_x^2 + p_y^2 + \omega^2 y^2 - \Lambda^2 x^2). \tag{31}$$

The steps are equidistant with a spacing $\hbar\omega$ and a width depending on the Lyapunov exponent Λ [21]. Representative conductance curves are shown in figure 3. Similar curves are also found in other geometries, e.g. see [22–24].

It was convenient for the calculation to assume a linear transition state, but since the semiclassical results only depend on properties invariant under canonical transformations (actions and stabilities) the above results are valid more generally: the position of the steps is given by the quantization of the action of the transition state and the width is determined by the Lyapunov exponent.

5. Magnetic fields

Magnetic fields perpendicular to the plane of the electron gas can also change the conductance. Because of the Lorentz force a Larmor precession is superimposed onto the linear motion so that the transition state will not usually be a straight line in position space; it will still be a periodic orbit. This is easiest to see for an analytically solvable case, which for a parabolic saddle which without magnetic field is described by

$$H = \frac{p_x^2 + p_y^2}{2m} - m\Lambda^2 x^2/2 + m\omega^2 y^2/2. \tag{32}$$

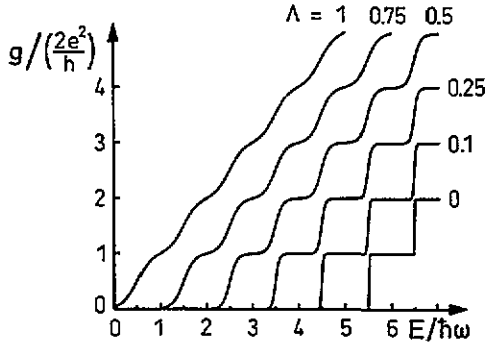

Figure 3. Conductance plotted against energy in a parabolic constriction for different Lyapunov exponents Λ. For better visibility the curves are displaced to the right, corresponding to a shift in the energy of the saddle.

Without a magnetic field the general motion is the superposition of an oscillation in y and an exponential escape in x. The transition state is obviously the motion in the y-direction. With magnetic field, motion can again be described as a superposition of an oscillation and an exponential runaway, but with different frequencies and new eigendirections in phase space.

The equations of motion for a particle of mass m and charge e in this potential and a magnetic field $(0, 0, b)$ are

$$\ddot{x} = -\omega_0\dot{y} + \Lambda^2 x \qquad \ddot{y} = \omega_0\dot{x} - \omega^2 y \tag{33}$$

where the magnetic field appears in the Larmor frequency $\omega_0 = eB/m$. The exponential ansatz $x(t) = e^{\lambda t}x_0$, and similarly for y, leads to the characteristic equation

$$(\lambda^2 - \Lambda^2)(\lambda^2 + \omega^2) + \lambda^2\omega_0^2 = 0 \tag{34}$$

with eigenvalues

$$\lambda_\pm^2 = -\Omega \pm \sqrt{\Omega^2 + \omega^2\Lambda^2} \tag{35}$$

where $\Omega = (\omega^2 + \omega_0^2 - \Lambda^2)/2$. Since λ_+^2 is always positive it describes an exponential runaway which approaches the x-motion for vanishing magnetic field. Similarly, the negative λ_-^2 describes an oscillatory solution which goes over into the y-motion in the field-free case. According to [25] it is always possible to perform a canoncial transformation to a normal form (32) with new frequencies $\Lambda' = \lambda_+$ und $\omega' = \sqrt{-\lambda_-^2}$. Thus, the field-free old topology is recovered and one can use the results of the preceding section to characterize the conductance. The spacing between two steps is given by $\Delta E = \hbar\omega'$ and their widths is $\hbar\Lambda'$. From this, it follows that the steps become sharper with increasing magnetic field both relative to the spacing (ΔE/widths $= \omega_-/\lambda_+ \to \infty$) and also in absolute terms (since $\lambda_+ \sim \omega\Lambda/\omega_0 \to 0$).

The normal form cannot be achieved by a point transformation alone, so that the periodic orbit of the transition state becomes an ellipse in configuration space (figure 4). However, the change in coordinates is still linear, so that it can be transferred to quantum mechanics; the quantum problem in a parabolic barrier in a magnetic field is exactly solvable [26].

In non-harmonic potentials the transition state has to be found numerically. In general, this will require a 2D search in initial conditions. However, if the potential still has a mirror symmetry across an axis, e.g. the x-axis $V(x, y) = V(x, -y)$, then one may again search in just one dimension, similar to the case without a magnetic field. Starting perpendicular to

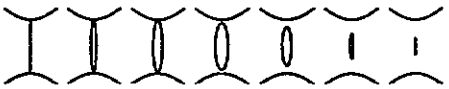

Figure 4. Transition state in a parabolic barrier for different magnetic fields (increasing from left to right). Without a magnetic field the PODS is a straight line connecting the two equipotential contours (left). For an increasing magnetic field it becomes elliptic (not touching the equipotential contour) and shrinks to a point for very large fields.

the x-axis, three coordinates are fixed (x_0, $y = 0$ and $p_x = 0$), the final one following from energy conservation. One now moves the initial condition until the trajectory returns exactly. This shows that the picture remains qualitatively unchanged unless the orbit bifurcates, in which case the flux calculated from this transition state is only an upper bound.

6. Concluding remarks

The models studied here are rather simple, with all classical trajectories either being reflected or smoothly crossing over the barrier. This allows us to evaluate the phase-space flux formula and the conductance solely in terms of classical quantities at the periodic orbit dividing surface and to include the tunnelling corrections. In more complicated cases the flux formula provides an upper bound. The tunnelling width may still be calculated but its relevance is less clear.

It is instructive to compare the present approach to others, in particular to the Landauer–Büttiker formula which relates transmission to scattering. There exists a well developed semiclassical theory for S-matrix elements which may be used to evaluate the conductance. The individual S-matrix elements are represented by superpositions of classical trajectories connecting ingoing and outgoing states. Via the Landauer–Büttiker formula, the conductance is obtained as the sums of the absolute values squared of suitably normalized S-matrix elements. This presentation allows for interference between contributions and is capable of producing conductance fluctuations. However, in the case of a single constriction the approach presented here is clearly superior in that all information is obtained from the transition state; moreover, tunnelling corrections can be included. It would be interesting to compare the two approaches quantitatively as this should shed light on the accuracy of the semiclassical approximation in general.

If the model is more complicated and the classical trajectories show backscattering, then the phase-space formula obviously only provides an upper bound on the flux, the reason being that not every point in the Poincaré surface of section at the transition state corresponds to a reactive trajectory. Numerical calculations for several systems indicate that the set of points leading to a reaction is a fat fractal [27]. The characteristics of a fat fractal are that it has a finite area S_0, but that this area is approached nonlinearly with the resolution ϵ of the boxes with which it is measured, i.e. $S(\epsilon)S_0 + S_1\epsilon^\beta$. The primitive semiclassical quantization which leads to the steps corresponds to a simple quantization of the area of the transition state, namely equation (25). If the area is a fat fractal then one can speculate that there will be nonlinear corrections to the average classical trend, just as in the formula for the density of states of a fractal drum [28,29]: if one assumes that Planck's constant h sets the scale with which the flux-carrying area is measured, then as a function of h at fixed energy the average conductance will be proportional to $S_0/h + S_1h^{\beta-1}$. Calculations for simple billiard geometries suggest that S_1/S_0 may be small and/or β may be close to one so that the second term is masked by the quantum fluctuations.

Acknowledgments

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