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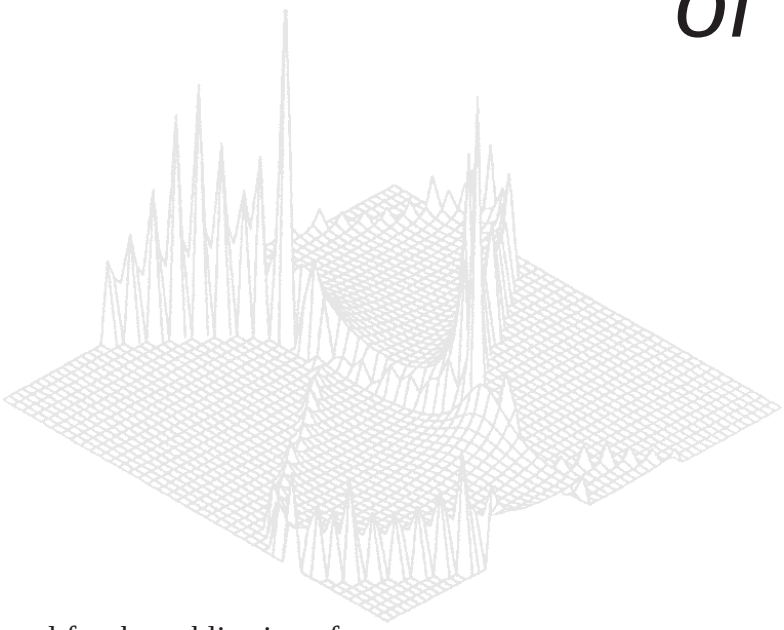

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Tunnelling of a Molecule

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Abstract

A quantum-mechanical description of tunnelling is presented for a one-dimensional system with internal oscillator degrees of freedom. The 'charged diatomic molecule' is frustrated on encountering a barrier potential by its centre of charge not being coincident with its centre of mass, resulting in transitions amongst internal states. In an adiabatic limit, the tunnelling of semiclassical coherent-like oscillator states is shown to exhibit the Hartman and Büttiker–Landauer times t_H and t_{BL}, with the time dependence of the coherent state parameter for the tunnelled state given by $\alpha(t) = \alpha\, e^{-i\omega(t+\Delta t)}$, $\Delta t = t_H - i t_{BL}$. A perturbation formalism is developed, whereby the exact transfer matrix can be expanded to any desired accuracy in a suitable limit. An 'intrinsic' time, based on the oscillator transition rate during tunnelling, transmission or reflection, is introduced. In simple situations the resulting intrinsic tunnelling time is shown to vanish to lowest order. In the general case a particular (nonzero) parametrisation is inferred, and its properties discussed in comparison with the literature on tunnelling times for both wavepackets and internal clocks.

1. Introduction

The phenomenon of tunnelling was recognised almost immediately after the inception of quantum mechanics, and issues of its physical implications have since provided a stimulus for many aspects of the debate on the interpretation of the theory as a whole. Beyond a philosophical interest, however, the ascription of a 'time' to particle tunnelling processes has immediate physical importance, in applications as diverse as cosmology, semiconductor technology, and Josephson junction devices (Büttiker and Landauer 1986). Recent experimental advances (Enders and Nimtz 1993*a*, 1993*b*; Steinberg *al.* 1993) leading to direct tunnelling 'time' measurements have underlined the importance of pursuing a better theoretical understanding of the process (Landauer and Martin 1994).

Abstract quantum mechanical analyses of tunnelling frequently derive from considerations of wavepackets, as promoted especially by Hartman (1962) (see also MacColl 1932). These derivations (MacColl 1932; Hartman 1962; Leavens and Aers 1989; Hauge *et al.* 1987) suffer problems of interpretation in connection with definitions of such notions as bandwidth, wavefront, signal, and pulse reshaping (Enders and Nimtz 1993*a*, 1993*b*; Landauer and Martin 1994). The analogy between the Schrödinger and Helmholz equations has been developed leading to equivalent discussions of tunnelling in the electromagnetic context (Martin

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and Landauer 1992), although unless single photons are involved (Steinberg *et al.* 1993), the wavepacket difficulties remain. Alternatively, tunnelling can be 'measured' by various types of quantum mechanical clocks involving the dynamics of internal degrees of freedom, such as those discussed originally in a spacetime context by Salecker and Wigner (1958) and subsequently used by several authors for tunnelling (Rybachenko 1967; Büttiker 1983; Leavens and McKinnon 1994). The elapsed time is taken from changes to the internal state of the system undergoing tunnelling (for example Larmor precession), in comparison with a similar noninteracting system. In contrast with the wavepacket case, the analysis requires only total energy eigenstates. In some limits, the Larmor time agrees with that deduced by analysis of wave tunnelling through a pulsating barrier (Büttiker and Landauer 1982), and also from considerations of modulated incident waves (Büttiker and Landauer 1986).

In this paper we report on aspects of the tunnelling time problem analysed from the viewpoint of a quantum mechanical oscillator clock (in 1 dimension). Thus we work with an infinite number of internal states, as against more recent versions invoking say a three position clockface (Leavens and McKinnon 1994) or a $2s+1$ state magnetic Larmor clock (Rybachenko 1967; Büttiker 1983). As a physical analogy, our model is that of a charged diatomic molecule with internal vibrational states. The tunnelling of this molecule is frustrated by its centre of charge not being coincident with its centre of mass, so that its interaction with a barrier potential incurs transitions amongst the oscillator modes (through the entangling of internal and centre-of-mass degrees of freedom in the boundary conditions) which can be used as a measure of elapsed time.

In Section 2 below, the analogue for the molecular system of the semiclassical wavepacket formalism is developed. The tunnelling of coherent-like oscillator states (but which are monoenergetic) is shown to exhibit both the Hartman and Büttiker–Landauer times t_H and t_{BL}, in that the time dependence of the coherent state parameter of the tunnelled state is $\alpha(t) = \alpha e^{-i\omega(t+\Delta t)}$, $\Delta t = t_H - it_{BL}$. To our knowledge this appealing interpretation of the much-discussed imaginary part of the tunnelling time has not been given before.

In Section 3 below, an 'intrinsic' oscillator interaction time t_I is defined. Motivated by the full solution for the simple case of reflection from an infinite wall (see also Appendix A), the intrinsic time (for reflection t_{Ir}, or tunnelling, t_{It}), compares the internal oscillator state after interaction to the effect of an effective evolution operator $\exp(-iH_{eff}t_I/\hbar)$ acting on the initial oscillator state (before the interaction). Specifically, we take $H_{eff} = V_0(a + a^\dagger)/\sqrt{2}$ for a rectangular barrier of height $V_0 = \hbar^2\mathbf{K}^2/2M$. A perturbation formalism is developed, which in simple cases yields $t_{It} = 0$ to lowest order. However, it is inferred in the general case that the intrinsic times are of the order $t_I \simeq Mka/\hbar\mathbf{K}^2$, in contrast to the well known (barrier thickness independent) Hartman time $t_H = 2M/\hbar k\kappa$ and the Büttiker–Landauer time $t_{BL} = Ma/\hbar\kappa$, for tunnelling through a barrier of thickness a at energy $E < V_0$, with $E = \hbar^2k^2/2M$ and $V_0 - E = \hbar^2\kappa^2/2M$.

Both the infinite wall solution, and the perturbative *ansatz*, are special cases of a general transfer matrix formalism for the problem, discussed in Appendix

B. In Section 4 below we conclude the paper with some further discussion and interpretation of our results.

2. Semiclassical Limits

Specifically, the Hamiltonian is

$$H = \frac{P_1^2}{2m_1} + \frac{P_2^2}{2m_2} + \tfrac{1}{2}k(X_1 - X_2)^2 + V_0\Theta(X^* + \tfrac{1}{2}a)\Theta(\tfrac{1}{2}a - X^*)\,,$$

where the molecule feels the barrier potential† via its charge, located at coordinate $X^* = \alpha X_1 + (1-\alpha)X_2$. In the centre-of-mass system we have

$$H = \frac{P^2}{2M} + \frac{p^2}{2m} + \tfrac{1}{2}m\omega^2 x^2 + V_0\Theta(X^* + \tfrac{1}{2}a)\Theta(\tfrac{1}{2}a - X^*)\,, \tag{1}$$

where the total mass $M = m_1 + m_2$ and the reduced mass $m = m_1 m_2/(m_1 + m_2)$ as usual. Below, we require the oscillator excitation energy $\hbar\omega$ to be small compared with the kinetic energy, and subsequently also take the effective size (the offset between the centre of charge and the centre of mass) small compared with the de Broglie wavelength. In terms of the oscillator length parameter $d = \sqrt{\hbar/m\omega}$ and the centre-of-mass wavenumber k, we have

$$\frac{\hbar^2}{md^2} \ll \frac{\hbar^2 k^2}{2M}, \quad \frac{\hbar^2}{M\alpha^2 d^2} \gg \frac{\hbar^2 k^2}{M}.$$

For simplicity we take $\alpha = 1$, so that m, M are in fact regarded as adjustable parameters.

As a first approach, as an analogue of the wavepacket formalism, let us consider the effect of tunnelling on a 'semiclassical' internal oscillator clock system incident on a barrier. Thus we take a coherent-like state $|k, \alpha\rangle$ where each internal component $|n\rangle$ comes with an appropriately modified centre-of-mass momentum $\hbar k_n$ with kinetic energy $E_n = E - (n + \frac{1}{2})\hbar\omega$, so that the total energy E is constant:

$$|k, \alpha\rangle = \sum_{n=0} \frac{\alpha^n}{\sqrt{n!}}\, |k_n\rangle \otimes |n\rangle$$

(where as usual the mean internal excitation is $N \propto |\alpha|^2$ with variance $|\alpha|$). Ignoring for the moment the effect of the boundary conditions on the molecular oscillator, or alternatively, in an adiabatic limit where the period is long compared with the interaction timescale ($\hbar\omega \ll E$) so that the oscillator internal state is unaffected, the reflected and transmitted amplitudes are entirely determined through their dependence on centre-of-mass kinetic energy E_n.

† The rectangular barrier potential is displayed here, but below we shall also discuss related cases such as step potentials, infinite walls and δ-functions.

Let the transmission coefficient be $T = \exp(-\eta + i\phi)$ for a very thick rectangular barrier where η is the exponential damping factor κa. Considering each internal component $|k_n\rangle \otimes |n\rangle$, we have reflection and transmission coefficients R_n, T_n say of the type

$$T_n \equiv T(E_n) = T(E^\circ - n\hbar\omega) \approx T(E^\circ)[1 + (\eta' - i\phi')n\hbar\omega]\,,$$

where $E^\circ = E - \frac{1}{2}\hbar\omega$ and the primes denote energy derivatives. Following Hartman (1962) we have $\hbar\phi' = t_H$, while from Büttiker and Landauer (1986) $\hbar\eta' = -t_{BL}$. The overlap of the time dependent transmitted state with $|n\rangle$ is thus

$$\alpha(t)^n T(E_n) \simeq \alpha^n T(E^\circ) \exp[-in\omega(t + \Delta t)],$$

where $\Delta t \equiv \hbar(\phi' - i\eta') = t_H - it_{BL}$. Thus the real and imaginary parts of Δt are the Hartman and Büttiker–Landauer times respectively (the minus sign in the imaginary shift here signifies that the parameter of the coherent state, α, has suffered an exponential decay by $\exp[-\omega t_{BL}]$), thus providing a further interpretation of tunnelling time as an entity with both real and imaginary parts (Rybachenko 1967).

3. Intrinsic Time

We now move from the semiclassical analysis to a full analysis of the system, beyond the adiabatic regime, and including internal transitions induced by the barrier interaction. In order to motivate a new definition of 'intrinsic' time we firstly give the complete solution for the (reflection) of the system from an infinitely high potential wall.

Let the system initially be in the oscillator ground state with wavenumber k_0, and consider reflected waves with wavenumber k_n and internal quantum number n such that

$$\frac{\hbar^2 {k_n}^2}{2M} + (n + \tfrac{1}{2})\hbar\omega = \frac{\hbar^2 {k_0}^2}{2M} + \tfrac{1}{2}\hbar\omega = E\,,$$

with some amplitude ρ_n, so that the full solution of the time-independent Schrödinger equation is

$$|\psi\rangle = \tilde{\alpha}_0 |k_0\rangle \otimes |0\rangle + \sum_{n=0}^{\infty} \tilde{\rho}_n |-k_n\rangle \otimes |n\rangle\,, \tag{2}$$

with corresponding wavefunction† $\langle X, u|\psi\rangle$ given by

$$\psi(X,u) = \alpha_0 e^{ik_0 X} H_0(u) e^{-\frac{1}{2}u^2} + \sum_{n=0}^{\infty} \rho_n e^{-ik_n X} H_n(u) e^{-\frac{1}{2}u^2}\,. \tag{3}$$

† Allowing for the normalisation of the function $H_n(u)$ we have $\tilde{\rho}_n = \rho_n \sqrt{2^n \pi^{\frac{1}{2}} n!}$.

Although there may only be a finite number of real values of k_n, there will be an infinite number of imaginary values (i.e. $k_n = -i\kappa_n$); these correspond to nonpropagating modes. If (see Section 2 above for the case $m_1 = m_2$) the potential barrier is at $X_1 = 0 = X + \frac{1}{2}du$ we have for the vanishing of ψ at $X_1 = 0$

$$0 = \alpha_0 e^{-\frac{1}{2}ik_0 du} H_0(u) e^{-\frac{1}{2}u^2} + \sum_{n=0}^{\infty} \rho_n e^{+\frac{1}{2}ik_n du} H_n(u) e^{-\frac{1}{2}u^2} . \tag{4}$$

Again, if $\hbar\omega \ll E_0$ we may set $k_n \approx k_0 \equiv k$ say, and introducing $\Delta = kd$, we have

$$e^{-i\Delta u} e^{-u^2} = \sum_{n=0}^{\infty} \left(\frac{-\rho_n}{\alpha_0} \right) H_n(u) e^{-u^2} , \tag{5}$$

whereupon $-\rho_n/\alpha_0 = (-i\Delta)^n/2^n n!$, or equivalently $\tilde{\rho}_n/\tilde{\rho}_0 = (-i\Delta)^n/\sqrt{2^n n!}$ follows from the usual generating function for Hermite polynomials (Gradshteyn and Ryzik 1965). In fact, equation (5) is simply the equivalent in the position representation of the operator expession

$$e^{(-i\Delta/\sqrt{2})(a+a^\dagger)}|0\rangle = e^{-\frac{1}{4}\Delta^2} \cdot e^{(-i\Delta/\sqrt{2})a^\dagger} \cdot e^{(-i\Delta/\sqrt{2})a}|0\rangle$$

$$\equiv e^{-\frac{1}{4}\Delta^2} \cdot \sum_{n=0}^{\infty} \frac{(-i\Delta/\sqrt{2})^n}{\sqrt{n!}} |n\rangle ,$$

from which we infer that the reflected state is in this case generated by an effective evolution operator $1 - i(\Delta/\sqrt{2})(a+a^\dagger)$. This provides a means of introducing an 'intrinsic' oscillator clock time for reflection, transmission or tunnelling, if we take a specific form for the effective interaction Hamiltonian H_{eff}. In conformity with the above, we therefore adopt $H_{eff} = V_0[(a + a^\dagger)/\sqrt{2}]$ for a rectangular barrier† of height V_0. In a perturbative approximation where the oscillator is incident on the barrier in the ground state, and the effect of the interaction is slight, we then define

$$t_{Ir} = \frac{i\hbar\sqrt{2}}{V_0} \frac{\tilde{\rho}_1}{\tilde{\rho}_0} = \frac{2i\hbar}{V_0} \frac{\rho_1}{\rho_0}, \tag{6}$$

and analogously for tunnelled (or transmitted) states $\tilde{\tau}_n|k_n\rangle \otimes |n\rangle$,

$$t_{It} = \frac{i\hbar\sqrt{2}}{V_0} \frac{\tilde{\tau}_1}{\tilde{\tau}_0} = \frac{2i\hbar}{V_0} \frac{\tau_1}{\tau_0}. \tag{7}$$

† Further discussion of the definition of H_{eff} is given in the concluding remarks below (see Section 4).

In the present, infinite wall case, the definition (7) is not applicable unless we invoke the limit $V_0 \to \infty$, in which case $t_{Ir} \to 0$ where from (5) above

$$t_{Ir} = \frac{\hbar\Delta}{V_0} = \frac{2Mkd}{\hbar \mathbf{K}^2}. \tag{8}$$

In fact, precisely this result† for t_{Ir} is found from a perturbative treatment of the rectangular barrier for finite V_0, as we now show. To this end, we consider the expansion of all quantities in the dimensionless parameters $\Delta_i = k_i d$ and introduce a collective total order $^{(k)}$ in all such Δ_i:

$$\rho_n = \rho_n^{(0)} + \rho_n^{(1)} + \rho_n^{(2)} + \cdots;$$
$$\tau_n = \tau_n^{(0)} + \tau_n^{(1)} + \tau_n^{(2)} + \cdots.$$

In Appendix B, a transfer matrix formalism for the problem is discussed, in which the perturbative regime is considered. As discussed there, we assert (for the oscillator initially in the ground state) that in fact $\rho_n = \mathrm{O}(\Delta^n)$ for each such coefficient, so that there is an expansion of the form

$$\rho_n = \rho_n^{(n)} + \rho_n^{(n+1)} + \rho_n^{(n+2)} + \cdots \tag{9}$$

and so on, which can be inserted in order to solve the boundary conditions systematically for the various contributions. This *ansatz* amounts to the statement that the equations for a step-wise constant potential, are lower-triangular in terms of elementary blocks such as

$$\begin{pmatrix} \alpha_0 \\ \rho_0 \end{pmatrix}, \begin{pmatrix} \alpha_1 \\ \rho_1 \end{pmatrix}, \begin{pmatrix} \alpha_2 \\ \rho_2 \end{pmatrix}, \cdots$$

in a propagating sector, and moreover (still for the oscillator initially in the ground state) that the equations for the zeroth order (ground state) amplitudes are those of the usual point particle.

In practice, for the rectangular barrier problem (see equation 1) in order to implement the definitions of intrinsic tunnelling and reflection times (6) and (7) it is sufficient to give a straightforward analysis of the boundary conditions to be satisfied by the solution of Schrödinger's equation in each of the three distinct regions of the potential, regarded as functions of u, and to be satisfied order by order in u:

$$\left\{ \begin{array}{cc} V_I(x) = 0, & x < -\frac{1}{2}a \\ V_{II}(x) = V_0, & -\frac{1}{2}a \leq x \leq \frac{1}{2}a \\ V_{III}(x) = 0, & x > \frac{1}{2}a \end{array} \right\}. \tag{10}$$

† The *same* result also emerges for t_{Ir} in the infinite wall case if the oscillator is initially in the internal state $|n\rangle$, provided the definition of t_{Ir} involves the ratios of the appropriate amplitudes $\tilde{\rho}_{n\pm1}$ with $\tilde{\rho}_n$ (see Appendix A).

These solutions are given by

$$\begin{aligned}
\psi_I &= \alpha_0 e^{ik_0X} H_0(u) e^{-\frac{1}{2}u^2} + \rho_0 e^{-ik_0X} H_0(u) e^{-\frac{1}{2}u^2} + \\
&\quad \rho_1 e^{-ik_1X} H_1(u) e^{-\frac{1}{2}u^2} + \rho_2 e^{-ik_2X} H_2(u) e^{-\frac{1}{2}u^2} + \mathrm{O}(3), \\
\psi_{II} &= \gamma_0 e^{\kappa_0X} H_0(u) e^{-\frac{1}{2}u^2} + \delta_0 e^{-\kappa_0X} H_0(u) e^{-\frac{1}{2}u^2} + \\
&\quad \gamma_1 e^{\kappa_1X} H_1(u) e^{-\frac{1}{2}u^2} + \delta_1 e^{-\kappa_1X} H_1(u) e^{-\frac{1}{2}u^2} + \\
&\quad \gamma_2 e^{\kappa_2X} H_2(u) e^{-\frac{1}{2}u^2} + \delta_2 e^{-\kappa_2X} H_2(u) e^{-\frac{1}{2}u^2} + \mathrm{O}(3), \\
\psi_{III} &= \tau_0 e^{ik_0X} H_0(u) e^{-\frac{1}{2}u^2} + \tau_1 e^{ik_1X} H_1(u) e^{-\frac{1}{2}u^2} + \\
&\quad \tau_2 e^{ik_2X} H_2(u) e^{-\frac{1}{2}u^2} + \mathrm{O}(3).
\end{aligned}$$

where we have taken an incident wave with the oscillator in the ground state (and amplitude $\alpha_0 = 1$) and moreover have anticipated (9) and an expansion to second order in u. The solution is required as usual to be continuous across the boundaries at $X_1 = \pm\frac{1}{2}a$; however, the derivatives which are continuous are with respect to variable X_1, namely $\frac{\partial}{\partial X_1} = \frac{1}{2}\frac{\partial}{\partial X} + \frac{1}{d}\frac{\partial}{\partial u}$. The six equations constructed using these boundary conditions may be expanded to second order in u and examined order by order to isolate expressions for the scattering coefficients (it is necessary to separate orders of u and Δ). The scattering coefficients at the first excitation state may then be given after a matrix inversion as

$$\begin{pmatrix} \rho_1^{(1)} \\ \tau_1^{(1)} \end{pmatrix} \simeq \frac{e^{-ik_0a}}{4(\kappa_0 - ik_0)^2} \begin{pmatrix} -(\kappa_1 - ik_1) & 2\kappa_1 e^{-\kappa_0 a} \\ -2\kappa_1 e^{-\kappa_0 a} & (\kappa_1 - ik_1) \end{pmatrix} \begin{pmatrix} \xi \\ \frac{2\kappa_0}{(\kappa_0 - ik_0)} \xi e^{-\kappa_0 a} \end{pmatrix} \mathbf{K}^2 \,, \quad (11)$$

where

$$\xi \equiv -\frac{2ik_0}{(\kappa_0 - ik_0)} \,, \quad (12)$$

and the standard solution for ρ_0 has been used. If we disregard components which are of order $e^{-2\kappa_0 a}$ in the limit where $\kappa a \gg 1$, and identify $k_i \equiv k$, $\kappa_i \equiv \kappa$ for $i = 0, 1$ then the reflection amplitude is evaluated from the first line of (11) as

$$\rho_1^{(1)} = \frac{ik}{2(\kappa - ik)^2} e^{-ika} \mathbf{K}^2 = \frac{i\Delta(\kappa + ik)}{2(\kappa - ik)} e^{-ika} \quad (13)$$

and so invoking the standard solution $\rho_0 = -[(\kappa + ik)/(\kappa - ik)]e^{-ika}$ in this limit we have

$$\frac{\rho_1^{(1)}}{\rho_0^{(0)}} = -\frac{i\Delta}{2} \,. \quad (14)$$

Therefore the reflection time is from (6)

$$t_{Ir} = \frac{2Mkd}{\hbar \mathbf{K}^2}\,, \tag{15}$$

in complete agreement with (8). Similarly, the transmission amplitude arises from the second line of (11). Strikingly, in the limit that the k_i and κ_i are identified, the second row of the matrix is orthogonal to the column vector involving ξ, which as $\tau_0^{(0)} \neq 0$ from the usual tunnelling analysis, implies

$$t_{It} \equiv 0\,. \tag{16}$$

Although it is tempting to infer from this analysis that the intrinsic tunnelling time t_{It} vanishes in general, the nature of the derivation and the validity of the answer (which after all only applies strictly in the limit $\hbar\omega \ll E$) implies that the result is a special case. Analysis of related problems (Bulte 1996) such as the step-up and step-down potentials, and the δ-function potential, confirms this, and suggests that a value for t_{It} of the same order as (15) is more reasonable. In Section 4 below, we give an extended discussion of these points and strengthen our conclusions as to the meaning of our oscillator calculations.

4. Conclusions

In this paper we have considered the quantum mechanical problem of the interaction of a system with internal oscillator degrees of freedom with an external barrier potential, considered as a model of the tunnelling of a charged diatomic molecule in one dimension. The system is frustrated by its centre of charge not being coincident with its centre of mass, so that the barrier interaction causes transitions between internal oscillator states. Based on this, an 'intrinsic' time for reflection, transmission or tunnelling is introduced and evaluated in simple cases. Here we discuss further the broader implications of these results for the general question of barrier interaction 'time'.

Firstly we should comment on the 'semiclassical' times involving coherent-like oscillator states incident on the barrier (Section 2). They are the analogue for the oscillator system of the usual wavepacket results, and reproduce the Hartman and Büttiker–Landauer times via a complex shift in the time dependence† of the coherent state parameter, $\Delta t = t_H - it_{BL}$. Although involving eigenstates of total energy, the fact that the answers depend in detail on the composition of the incident oscillator state suggests that these results, and the equivalent analyses in the non-oscillator context leading to the promotion of t_H or t_{BL} as good measures of barrier interaction times, should be treated with caution, in favour of the 'intrinsic' time approach (Section 3).

For this we work in a perturbative context in which the effect of the interaction on the internal state of the system is slight. Thus, for example, there are only

† In fact Δt in this case represents an advance rather than a delay. However, different types of coherent-like states, for example with constant excitation energy rather than constant total energy, will manifest Δt as a delay time.

small amplitudes for the system to be found in neighbouring levels, if it is incident in a definite internal oscillator state. This reasonable assumption corresponds to taking the system to be almost point-like, so that the mis-match between the centre of charge and centre of mass is minimal. This is implemented in practice in the limit that the de Broglie wavelength of the system is much greater than the oscillator size, or $kd \ll 1$ for wavenumber k and size d. In order to obtain easily interpretable results, it is further convenient to neglect the oscillator excitation energy in comparison with typical centre of mass energies, $\hbar\omega \ll E$ and $\hbar\omega \ll V_0 - E$. Although these two regimes are not strictly compatible given the constraints on d, ω, and k, E, together with the reduced mass m and the total mass M, it is always possible to adjust the position of the centre of charge X^* (see equation 1 and Section 2) to improve the point-like assumption (d is effectively replaced by ϵd with a free parameter $\epsilon \ll 1$), so that we have accepted both limits for the purposes of the analysis.

It will be noticed that our calculations inevitably result in intrinsic times t_I proportional to the oscillator length d, without reference to the barrier size a. Again, it is reasonable to relate this to the fact that the interaction causing the oscillator transitions occurs only at the boundary of the barrier, over a distance d measuring the size of the oscillator†: Once the molecule is well inside the barrier, the problem reverts to that of the centre-of-mass behaviour. However, it is easy to envisage models wherein the system is 'continuously' frustrated inside the barrier (for example, linearly rising potentials could be treated exactly with the appropriate Airy functions). Our analysis is intended to provide definitions of intrinsic times valid for general tunnelling problems, and without going further into more complicated variations, we infer, on the basis of our calculations so far, that the parameter d should be replaced (up to geometrical factors) simply by the full barrier width a in general.

Although H_{eff} in Section 3 was motivated by the analysis of the infinite wall problem (see also Appendix A), it is straightforward to gain direct insight into its origin by expanding the interaction $V(X^*)$ into a centre-of-mass term and further (internal) contributions. Expanding $X^* = X_1 = X + \frac{1}{2}du$ (for $m_1 = m_2$),

$$V(X^*) = V(X) + \tfrac{1}{2}duV'(X) + \cdots ,$$

we recognise $u = \sqrt{\frac{1}{2}}(a + a^\dagger)$. Moreover, if V is stepwise constant (a combination of Heaviside Θ-functions, say, with height parameters V_0), then the derivative will involve (a sum of) δ-functions in X, whose matrix elements between centre-of-mass wavefunctions in perturbation theory must contribute further terms which can only involve functions of the appropriate wavenumbers, with overall dimension of inverse length. These general considerations are fully borne out by our actual calculations and justify the form of H_{eff} which we have adopted.

† Significantly, our calculated reflection time t_{Ir} for the infinite wall problem is independent of the initial oscillator state (see Appendix A). An interpretation of this is that, despite the oscillator in state $|n\rangle$ being more spread than in the ground state $|0\rangle$, the effective interaction over this distance is larger, in such a way that the two effects compensate one another, and a single, consistent intrinsic time emerges from the algebra.

Taking into account the above remarks, and the results of our calculations in Section 3, we conclude that the appropriate intrinsic times t_I based on the oscillator method are, for rectangular barriers of width a,

$$t_I \sim t_{Ir} \sim t_{It} = \frac{Mka}{\hbar \mathbf{K}^2} \tag{17}$$

for both reflection and tunnelling. This result is comparable to the Büttiker–Landauer (1982) and some of the Larmor (Büttiker 1983) forms in being proportional to the barrier thickness. However, in contrast to $t_{BL} = Ma/\hbar\kappa$, equation (17) vanishes near the bottom of the barrier, and rises to the 'physical' value of $Ma/\hbar\mathbf{K}$ (that is, the barrier width divided by the centre-of-mass speed) at the top of the barrier (rather than diverging, as in the Büttiker–Landauer case).

In considering further applications of the intrinsic time, the extension to the Dirac equation naturally arises (Leavens and McKinnon 1994; Davies 1986). It is possible that the 'visceral' nature of our oscillator clock, with its boundary interaction mechanism, may provide a robust way of retrieving our nonrelativistic results as a limit of a more complete relativistic analysis. As far as the nonrelativistic model itself is concerned, it would also be interesting, given the degenerate states $|k_n\rangle \otimes |n\rangle$, to analyse the full (infinite-dimensional?) symmetry algebra of the two dimensional problem, and to formulate the scattering theory in this context.

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Appendix A: Infinite Wall with Arbitrary Oscillator States

In Section 3 we considered the interaction of the molecular oscillator, initially in the ground state, with an infinite reflecting wall. From this the definitions (6) and (7) of intrinsic time were derived. Here we extend the analysis to the case of an initial oscillator state $|n\rangle$ of arbitrary level. In the notation of (3) we have, implementing† the vanishing of the wavefunction at $X_1 = 0$,

$$0 = \alpha_n e^{-\frac{1}{2}ik_n du} H_n(u) e^{-\frac{1}{2}u^2} + \sum_{m=0}^{\infty} \rho_m e^{+\frac{1}{2}ik_m du} H_m(u) e^{-\frac{1}{2}u^2} \tag{18}$$

or, setting again‡ $k_m = k_n \equiv k$, we have

$$2^m \sqrt{\pi} m! \rho_m = -\alpha_n L_{mn}(-\tfrac{1}{2}i\Delta)\,, \tag{19}$$

where we have introduced (Gradshteyn and Ryzik 1965)

$$\begin{aligned} L_{m_1 m_2}(z) &= \int_{-\infty}^{+\infty} du e^{-u^2} H_k(u+z) H_l(u+z) \\ &\equiv 2^{(M/2)} \pi^{\frac{1}{2}} m! z^{(M-m)} L_m^{(M-m)}(-2z^2)\,, \end{aligned} \tag{20}$$

where $m = \min(m_1, m_2)$, $M = \max(m_1, m_2)$ and $L_m^{(n)}$ is an associated Laguerre polynomial.

In the case where the interaction is a small perturbation, we expect that the reflected molecule will predominantly be in the oscillation state $|n\rangle$ with some amplitudes to be found in states $|n \pm 1\rangle$. In the present case use of (20) leads to $\rho_{n-1} = in\Delta\alpha_n$, $\rho_{n+1} = \frac{1}{2}i\Delta\alpha_n$ or in view of the normalisation (see Section 3),

$$\tilde{\rho}_{n+1}/\tilde{\rho}_n = -i\Delta\sqrt{\frac{(n+1)}{2}}, \quad \tilde{\rho}_{n-1}/\tilde{\rho}_n = -i\Delta\sqrt{\frac{n}{2}}\,. \tag{21}$$

If we equate these ratios with (off-diagonal) matrix elements of the effective unitary evolution operator $1 - i\frac{V_0 t_{Ir}}{\hbar}\frac{(a+a^\dagger)}{\sqrt{2}}$ as in Section 3 [see for example (6)], then the derived intrinsic reflection time is precisely in accordance with our previous values (8) and (15).

Appendix B: Transfer Matrix Formalism

From an abstract point of view the scattering problem should be viewed in terms of a transfer matrix which relates data defining the system between different sectors (of a piecewise constant potential, say). As an illustration we take a generic propagating sector with amplitudes α_n, ρ_n and discuss the boundary conditions

† As mentioned above, the sum over m must revert to nonpropagating modes for $m \geq E^\circ/\hbar\omega$. However, as our expansion in Δ is to lowest order, we ignore this inessential complication to the notation.

‡ This limit is not necessary for the general solution.

satisfied by the solution (for convenience, at $X_1 = 0$). The generalisation of (18) together with (20) now yields for $m = 0, 1, 2, \cdots$

$$\sum_{n=0}^{\infty} e^{-(\Delta_n^2/16)} L^-_{mn} \alpha_n + e^{-(\Delta_n^2/16)} L^+_{mn} \rho_n = \chi_m \,, \tag{22}$$

where the superscript $(\pm)$ here refers to the L matrices accompanying the nth coefficient evaluated at argument $(\pm i\Delta_n/4)$, and the right-hand sides are taken as input for this sector. Similarly, evaluating the derivative $(\partial/\partial X_1)$ at $X_1 = 0$, and using standard Hermite function recurrence relations, the continuity equation becomes

$$\sum_{n=0}^{\infty} e^{-(\Delta_n^2/16)} [i\Delta_n L^-_{mn} + nL^-_{m(n-1)} - \tfrac{1}{2} L^-_{m(n+1)}] \alpha_n + e^{-(\Delta_n^2/16)} [-i\Delta_n L^+_{mn} + nL^+_{m(n-1)} - \tfrac{1}{2} L^+_{m(n+1)}] \rho_n = \phi_m \,. \tag{23}$$

Consider a system of the generic type

$$\sum_{n=0}^{\infty} M_{mn} \lambda_n = \mu_m \,,$$

where quantities depend on a parameter ϵ such that $M_{mn} = \mathrm{O}(\epsilon^{|m-n|})$. If coefficients λ, μ are such that $\lambda_n = \mathrm{O}(\epsilon^n)$, that is

$$\lambda_n = \lambda_n^{(n)} \epsilon^n + \lambda_n^{(n+1)} \epsilon^{n+1} + \cdots ,$$

then the m equation is likewise $\mathrm{O}(\epsilon^m)$, for example

$$[M^0_{mm} \lambda_m^{(m)} + M^{(1)}_{m(m-1)} \lambda_{m-1}^{(m-1)} + \cdots M^{(m)}_{m0} \lambda_0^{(0)}] \epsilon^m + \cdots = \mu_m^{(m)} \epsilon^m + \cdots .$$

Thus the system of equations for $\lambda_0^{(0)}, \lambda_1^{(1)}, \lambda_2^{(2)}, \cdots$ is in lower triangular form, and indeed the higher order contributions $\lambda_m^{(m+1)}, \lambda_m^{(m+2)}, \cdots$ can also be so ordered.

In the present case (22), (23) the matrix equivalent to M_{mn} has 2×2 block form in terms of L_{mn} and $L_{m(n\pm1)}$. However, the same general structure in terms of the parameters Δ_n is observed [cf. especially (20)], so that the system becomes essentially lower-triangular in terms of elementary blocks such as

$$\begin{pmatrix} \alpha_0^{(0)} \\ \rho_0^{(0)} \end{pmatrix}, \begin{pmatrix} \alpha_1^{(1)} \\ \rho_1^{(1)} \end{pmatrix}, \begin{pmatrix} \alpha_2^{(2)} \\ \rho_2^{(2)} \end{pmatrix}, \ldots .$$

As explained in Section 3, however, for the elementary analysis required in order to extract intrinsic times in simple cases, an explicit expansion in powers of u and Δ suffices.

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